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# Lawrence Livermore National Laboratory

September 8, 1989

Dr. William Woodward  
Energy Research Advisory Board  
U.S. Department of Energy  
Washington, D.C.

Dear Dr. Woodard,

Please find enclosed details of the Lawrence Livermore National Laboratory Cold Fusion experiment titled "Cold Fusion Electrolytic Experiments" by Aldridge, Contolini, Ishikawa, and Slaughter of LLNL. In their experiments a sufficiently large number of parameters were measured to meet the requests contained in Dr. Huizenga's letter of August 9, 1989. This experiment was briefly described in the original summary of the LLNL Cold Fusion Program from our Director John Nuckolls to the Department of Energy on June 16, 1989. See page two, paragraph two. Please feel free to call the authors or myself for additional information.

Sincerely,

John F. Holzrichter  
Assistant to the Director  
for IR&D

cc: J. Nuckolls  
J. Bigeleisen

attachments:

Cold Fusion Electrolyte Experiments  
Summary - LLNL Cold Fusion Experiments

## Cold Fusion Electrolytic Experiments

*F. T. Aldridge, R. J. Contolini, M. Y. Ishikawa and D. R. Slaughter*

This is a summary of 'O' division attempts to duplicate the Fleischmann-Pons electrolysis cold fusion experiments. Two types of cells were used, one a duplicate of the Fleischmann-Pons cell and another with a much larger flat plate electrode.

The first experiments were run in a duplicate of the Fleischmann-Pons cell, with somewhat smaller dimensions. The cathode was a central Pd cylinder of 6.5mm diameter and 70mm length. Only about 55mm of the Pd cylinder was immersed in the solution. The anode was a Pt coil wound on glass rods which were held by teflon end spacers. The teflon spacers also held the central Pd cathode. The electrode assembly was mounted inside a 24mm inside diameter graduated cylinder. The cell contained 24 cc of 0.1 N. LiOH in 99.8% D<sub>2</sub>O.

The first run was made at 4 amperes and 12 volts, with a current density of 0.4 amperes per square centimeter on the Pd electrode. After about 1.3 hours, the Pd rod suddenly glowed red, then white for a brief moment, and the cell exploded with a force similar to a small firecracker. The Pd electrode was discolored, except where it had been covered by the teflon holder. The teflon was not distorted or melted where it had been in contact with the Pd rod. Most of the force of the explosion was probably due to the recombination of electrolytically produced hydrogen and oxygen which was ignited by the hot Pd surface. The discolored layer was 0.5 micron thick and contained Na, K, Ca, and Li. All the metals except Li probably were leached out of the glass vessel by the action of the LiOH solution.

The explosion was possibly a surface reaction not involving the bulk of the Pd rod, or a reaction between the electrolyte or hydrided Pd and a Li-Pd alloy formed on the electrode surface. This may be the same phenomena reported by Fleischmann and Pons.

The apparatus was rebuilt and more experiments run without explosions. In these experiments, current densities were used of 0.21 to 0.39 amperes per square centimeter for periods of up to 32 hours. A NE213 Nuclear Enterprise organic scintillator was used to detect both neutrons and gamma rays. Neither neutron nor gamma counts were seen above the background level.

A water jacketed cell was used in an attempt to measure cell heat output by measuring the temperature of the water entering and leaving the water jacket. The system was too crude to give the accuracy of interest and the attempt was abandoned. Accurate thermal

measurements on this system will require a great deal of careful design work or the purchase of a commercial calorimetry system if valid results are to be obtained. Also, a closed system will be needed, where the hydrogen and oxygen produced are catalytically recombined within the cell and no gas exchange is allowed with the surroundings. (The Fleischmann-Pons type of experiments, which are not closed in this sense, have been widely criticized for their "open" feature.)

Some of our experiments were run in a larger cell. The Pd cathode used was a ten centimeter square plate 0.5 millimeters thick. Two ten centimeter square Pt mesh anodes were spaced one half inch from each side of the cathode. The electrodes were held in place with teflon spacers and the electrode assembly was placed in a glass vessel. This cell was placed in a cooling bath and the electrolyte temperature was approximately 40°C during the run. The cell was run for 65 hours at 10 amperes and 18 volts, with a current density of 0.05 amperes per square centimeter. A NE213 Nuclear Enterprises organic scintillator detector was used to detect gamma rays and neutrons. Several periods of enhanced neutron + gamma emission were seen at levels of 4 to 7 times the background level. A building wide power failure, caused by a bird shorting a power line, ended the experiment an hour after these observations. When the experiment was restarted after power was restored no more enhanced emission episodes were seen.

Since the enhanced count levels could not be duplicated, we do not know if they were significant. They may have been caused by unknown variability in detector sensitivity.

After the experiment was concluded, the electrolyte and the Pd electrode were analyzed for possible fusion products. The Pd electrode showed no  $^3\text{He}$  or  $^4\text{He}$  above the expected background levels. The electrolyte showed a 13% enrichment in tritium, but this was essentially what would be expected from the electrolytic enrichment of the  $\text{D}_2\text{O}$ , using literature values for enrichment factors.

The initial volume of electrolyte in the cell was 1350 ml. During the course of the experiment, 560 ml of electrolyte was added to keep the cell volume constant, for a total volume of 1910 ml of electrolyte used. The total current passed through the cell would have electrolyzed 220 ml of electrolyte; 340 ml was lost as vapor or spray entrained in gases liberated by electrolysis. The hydrogen produced by electrolysis would be depleted of tritium. This tritium would remain in the electrolyte, while the vapor or spray losses would not be isotopically selective. Therefore, the remaining electrolyte should be tritium enriched by a factor of  $\sim 1.130$ .

Analysis of the electrolyte showed 57.1 picocuries of tritium per ml before the experiment and 63.6 picocuries per ml after the experiment for an enrichment factor of 1.128. Thus, no

tritium production due to cold fusion was seen. The D<sub>2</sub>O used was obtained from Sigma Chemical Company and has an isotopic purity of 99.8% deuterium.

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June 16, 1989

## LLNL REPORT ON COLD FUSION

With the appearance of reports on "Cold Fusion", scientists at the Lawrence Livermore National Laboratory began an increasingly sophisticated series of experiments and calculations to explain these phenomena. These experiments can be categorized as follows: (a) simple experiments to replicate the Utah results, (b) more sophisticated experiments to place lower bounds on the generation of heat, and production of nuclear products, (c) a collaboration with Texas A&M to analyze electrodes and electrolytes for fusion by-products in a cell producing 10% excess heat (we found no by-products), and (d) attempts to replicate the Frascati experiment that first found neutron bursts when high pressure deuterium gas in a cylinder with Ti chips is temperature cycled.

We failed in items (a) and (b) to replicate either the Pons/Fleischmann or Jones phenomena, and appear to have duplicated the Frascati results, (d). Summaries of (b) through (d) are described below, as is a theory effort based on cosmic ray muons.

### Electrolytic Cells

A typical experiment involved comparisons of electrolytic cells using  $D_2O$  and 0.1 M LiOD with cells using  $H_2O$  and 0.1 M LiOH. Palladium wires 1 mm in diameter were used in the two runs described below, the first were simply swaged and drawn, but since there were reports that remelting was necessary we annealed the wire for the second run for 3 hours at  $600^\circ C$ . The first pair of cells ran for 156 hours in a common bath, each with a 35 cm long swaged wire, with current densities starting at  $90 \text{ mA/cm}^2$ , increasing to  $270 \text{ mA/cm}^2$ , and then to  $450 \text{ mA/cm}^2$ . The slight temperature difference between the two cells remained essentially constant throughout the run, that is there was no excess heat difference between them. Using relatively low efficiency neutron detectors, no neutrons above background were observed. An analysis of the Pd for  $^3\text{He}$  and  $^4\text{He}$  was negative, setting limits of a few times  $10^{10}$  atoms per gm. After cutting a length of this wire from the above  $D_2O$  cell it was run in another thin-walled cell for 4 more days with a Geiger counter near it. No counts above background were observed. A second experiment used a pair of cells with annealed Pd, and it ran for 330 hours at a current density of  $230 \text{ mA/cm}^2$  with similar null results. Analysis of that wire showed no helium buildup either. In both cases the thermometry might have missed seeing excess heat at the 10% level.

One cell similarly prepared to the one described above was setup for sensitive neutron counting. The cell ran for 167 hours and featured a proton recoil telescope detector that eliminated all background counts, and it used a sensitive liquid scintillator (7% efficiency, absolutely calibrated, with neutron/gamma pulse shape discrimination). The latter set a limit of 0.2 neutrons/sec from the 27 g Pd wire, half the level reported by Jones at BYU with a 5 g sample.

Several other experiments with electrolytic cells were run in a second independent group. Their experiment was designed to test for helium buildup in Pd, for neutrons, and for excess heat. Calorimetry with a minimum thermal detectivity of about 2% of the input electrical power was used. Similar (null) results were found by this group. If the excess heat is a 10% effect, as observed at Texas A&M, then this calorimeter has sufficient sensitivity and no effect was observed.

#### Collaboration with Texas A&M

We analyzed samples of Pd from a cell run at Texas A&M by Srinivasan, who reported 340 mW heat flow into their sensitive microcalorimeter with 300 mW heat input, indicating an excess heat generation of 40 mW. However, it should be remembered that an additional power input of 150 mW was needed to separate the D<sub>2</sub>O molecules. To the level of  $3 \times 10^5$  <sup>3</sup>He atoms and  $5 \times 10^8$  <sup>4</sup>He atoms in the sample (about 20 mg) there was no helium generation in the wire. This is many orders of magnitude below the level associated with 40 mW of fusion power for one hundred hours. Surface analysis with Auger and SIMS techniques revealed a substantial layer of material (Ca, C, Fe, O, Cl, Li, N, Cu and traces of other elements) plated out on the surface. These gave no clue as to possible excess heat generating mechanisms however they indicate suspicious additional electrochemical mechanisms. An analysis of the electrolyte before and after the run showed no change in the tritium level in the original electrolyte.

#### Neutron Burst Experiments

Experiments using pressurized deuterium cylinders loaded with Ti in various forms, similar to the Frascati experiments, were carried out by several groups at Livermore. In a large number of runs, cycling the temperature from 2N to room temperature, the results were null. Very recently, with detector efficiency improved to 15%, with higher pressure operation to 60 Atm, and with a pair of coincidence detectors incorporating burst mode triggering in the detection circuits, one group observed what appear to be bursts of a few hundred neutrons. These do not correlate with the temperature of the cylinder at the time of the bursts, which is contrary to data from LANL. Consistent with Sandia, it requires multiple detectors and the observation of coincident signals to place some confidence in the results. Many

spurious signals appear on one detector but not another, and this could be misinterpreted as neutrons. At present, the temporal signals are being studied to assure ourselves that the signals are indeed neutrons. Plans to repeat this in DT gas have been made.

#### Muon Theory

A theory to explain that the fusion neutrons observed by Jones etc. were generated from cosmic muons was developed here. The theory falls short by about two orders of magnitude to explain the level of neutrons seen by Jones. It was found that charge exchange with deeply trapped electrons around the Pd (or Ti) limits the muon to catalyzing at most a dozen events. In 13% of the muon catalyzed D-D reactions the muon sticks to the resulting He atom, and must subsequently be reionized by deeply trapped (a few keV) electrons. Instead, however, the muon charge exchanges. Further, experiments in Japan with accelerator generated muons, reported recently in Santa Fe, definitively set an even lower limit to muon catalyzed reactions than the above theoretical explanation.

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September 14, 1989

Dr. William Woodard  
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ER-6 3F-061  
Office of Energy Research  
1000 Independence Avenue, SW  
Washington, DC 20077-9381

Dear Dr. Woodard:

As requested, the Oak Ridge National Laboratory is providing the enclosed reports summarizing the four separate "cold fusion" experiments underway at the Laboratory. Like many groups around the world these experimenters at times observed some unusual and provoking events. However, we find no compelling observations to support the phenomena of nuclear events leading to "cold fusion". Anomalous observations persist in some of our experiments as detailed in the enclosed reports and we reserve interpretation of these until more careful measurements can be made to explain the causes.

I hope that these reports will be of value to you in your investigation. If you need further information or have any questions please call me.

Sincerely,



Bill R. Appleton

BRA:aj

Enclosures

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ELECTROCHEMICALLY INDUCED REACTIONS OF  
HYDROGEN ISOTOPES WITH METALS

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## ABSTRACT

The Metals and Ceramics Division of the Oak Ridge National Laboratory has, under the direction of the United States Department of Energy, undertaken an exploratory investigation of the phenomena occurring when hydrogen isotopes are electrolytically introduced into palladium and other metals. Major efforts were expended for the study of the deuterium isotope into palladium to better understand the touted "cold fusion" that has been proposed recently. Neutron counting analyses were inconclusive, showing a few unexplained events during electrolysis. In these instances neutrons were measured for short periods of time near operating electrolytic cells. Gamma ray detection showed no response attributable to fusion events. Tritium counts were noted to rise 5-7 fold for a short time in one cell with a gold anode and a palladium cathode. No conclusive evidence has yet been acquired to indicate excess energy production in our two calorimetric experiments undertaken prior to this writing.

Extensive analyses of electrode materials reveal that the absorption/desorption of deuterium into palladium restructures the metal appreciably. After extensive electrocharging (more than 0.85 D/Pd) the imbibed material is slowly lost (ie. more than 0.6 D/Pd after 700+ hours of exposure to laboratory environment. The concentration of deuterium is measured by four different techniques and the defect structure of the palladium is very extensive as measured by multiple techniques. With the number of extraneous phenomena that accompany electrocharging, one is cautioned to note that there are several energetic processes involved. This progress report details the approach used in evaluating the experimental and theoretical aspects of electrolytic introduction of hydrogen isotopes into palladium. No definite conclusions are presented concerning "cold fusion" since the details of the processes are not defined well enough.

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## I. INTRODUCTION

Few phenomena in science have raised as much interest in the last decade as has the "cold fusion" as an explanation of the events occurring when hydrogen isotopes (primarily deuterium) are electrochemically charged into palladium and other metals. This report is the result of a multidisciplinary experimental and theoretical evaluation of the processes that are involved. The approach is maintained at a very conservative level, both in expenditures and in scientific evaluation. Regardless of the final outcome of this research, the impact on the scientific community can be beneficial in several aspects. First, a complete review of many of the fundamental concepts is mandated--the proponents of the process are not neophytes, novices, or publicity seekers. Second, the vast number of unexplained phenomena require learned analyses and experimental verification. Third, a complete understanding of the process necessarily involves multidisciplinary communication and experimentation. Fourth, theoretical analyses are forced to explore areas that would not have been investigated without the impetus provided by the recent postulates. In many instances we are asked to question the very bases of our concepts and training. In any case, we cannot disregard any promise of an infinite supply of energy at a cost that is potentially very low.

This is an interim report as the fiscal year 1989 comes to a close.

## II. CALORIMETRIC ANALYSES FOR COLD FUSION PROJECT

### A. OVERVIEW

Calorimetric analyses are of prime importance in the studies of electrochemical fusion as proposed by B. Stanley Pons and Martin Fleischmann<sup>1</sup>. There are several sources of heat within electrochemical cells and each must be considered in a complete accounting of heat balances. For reference purposes the cell is considered to be of standard design with three electrodes: (1) A working electrode of an inert metal (platinum in most instances) that serves as the anode and establishes the driving voltage for current transfer to the (2) cathode (palladium in this instance, although titanium and other metals are prime candidates) where the reduction to form hydrogen occurs) and (3) a reference electrode that allows measurement of the electrode potential per se. In normal electrolysis the cell is configured as above and the anode is also inert. However in the current studies the palladium (or other candidate metal) electrode is reactive and hydrogen is incorporated into the metal matrix to ostensibly form the precursors for cold fusion (at least in the case of the deuterium isotope).

Relevant thermochemical data are given in reference 2:

	H <sub>2</sub> O	HDO	D <sub>2</sub> O	
H°(FORMATION) (g)	-57.7976	-58.735	-59.5628	Kcal/mol
(l)	-68.3171	-69.393	-70.4133	
Go(FORMATION) (g)	-54.6352	-55.828	-56.0670	Kcal/mol
(l)	-56.6902	-57.926	-58.2062	
S°	(g) 45.105	47.66	47.379	eu
	(l) 16.716	18.95	18.162	

These data are of prime interest in calorimetric analyses of electrochemical processes and will serve as reference for subsequent discussions.

### B. CALORIMETER DESIGN

#### 1. REACTION VESSELS

Electrochemical cells are placed in a constant temperature bath with an interspersed medium tailored to give the desired thermal transfer modulus. A typical design is given in Fig. 1, showing a well defined air gap between the cell per se and the surrounding environment. The size of the gap and/or the medium within the gap can be modified for evaluation of the heat generated within

the internal cell. Cell and jacket sizes vary depending upon the size of the electrodes. Most have a capacity of 75 cc up to 150 cc. Teflon was used for the lid to the pyrex glass cells because of its hydrophobic character, chemical inertness, low thermal conductivity and machinability. The isoperibolic behavior is trimmed to produce the desired temperature rise within the cell. A trade-off is met to assure adequate precision without an undue temperature rise. Each was calibrated individually as outlined below.

Each cell contains the electrodes used for electrochemical analyses, the thermistor temperature sensor, a calibration heater and a stirrer.

## 2. TEMPERATURE SENSORS

The primary sensors used in this study are negative temperature coefficient thermistors with pyrex glass encapsulation to give a response time of ca. 6 seconds. An electronic comparative circuit was used to measure the temperature difference between the electrolyte solution in the calorimetric cell and the surrounding constant temperature bath. Vigorous stirring of the 35 gallon water bath was carried out with a speed controlled (1500 rpm to 2000 rpm: 55 gallons per minute) stirrer. This bath temperature was maintained to  $\pm 0.002$  deg. C by a triple action (proportional control, optimized rate of approach, and reset), using the absolute amplified output of the reference thermistor circuit. This method of control is monitored by a separate thermistor circuit and has been found to control to within the 0.005 deg. C for months. Short duration deviations were noted for perturbations such as adding new cells and/or adding water to refill the bath. Such excursions were of short duration (2-5 min.). The bath temperature was continuously monitored, simultaneously with the monitoring of the eight temperature channels, cell currents, and cell voltages for each of the eight cells. The use of the differential mode of measuring the temperature difference between each cell and the reference bath temperature further minimizes errors that would occur if absolute cell temperatures were employed. All measurements were made with the bath temperature at the  $25 \pm 0.01$  deg C normally used as the thermochemical reference state.

A dedicated personal computer was used so that each variable was measured, plotted, and recorded every 30 seconds around the clock.

## 3. CALIBRATION HEATERS

Each cell is equipped with its own calibration heater. These were nichrome resistance wires (10 to 15 ohms) bifilar wound on

sintered alumina supports and subsequently coated with a thin layer of cured epoxy resin. Each was connected via copper leads to form a 4-lead (current and voltage) connection more than 1.5 inches below the electrolyte level in the cell. In this manner, one is assured that the heat generated in the nichrome is dissipated within the reaction medium and that the current (I) and voltage (V) measurements accurately define the heat ( $E \cdot I$ ).

#### 4. STIRRERS

Stirrers were fabricated for each of the cells since homogeneity was not achieved with the normal hydrogen and oxygen bubbling accompanying current densities of 10 to 500 milliamps per square centimeter of electrode surface. This was noted in both temperature measurements and in the distribution of bubbles. With stirring, the bubbles were homogeneously distributed (a phenomenon which correlated well with temperature homogeneity). Each stirrer was a 30 degree pitch thin wall (ca. 0.2 mm) spiral. This design proved to give efficient stirring with much less heat generation than conventional bladed propellers (at a much slower rotational speed).

The stirring process also serves to assure that each bubble has equilibrated with the water vapor in the electrolyte. In this manner one is assured that full saturation is achieved prior to release into the headspace above the electrolyte. This is important in the heat balance, as discussed below, for an operating electrolytic cell. Without stirring, oxygen and hydrogen gas generated near the top of the liquid do not have time to become saturated with water vapor. Stirring recycles each bubble several times and water vapor saturation is assured.

#### 5. ELECTRICAL CALIBRATION

Two complementary methods of calibration of the cells were employed:

a. Preliminary calibrations were performed with each cell equipped with a full complement of electrodes and electrolyte prior to initiation of the electrochemical processes. Two examples are given, one with a 1/2 inch jacketing layer of water around the cell and a second with only the 2 mm glass wall as a heat conduction path to the constant temperature bath. As can be seen in Fig. 3 and 4, respectively, there is a marked difference in response (temperature rise for a given energy input). The nonlinear response of the thermistors is noted in the response over the greater temperature range of the jacketed cell (the dashed line of Fig. 3 is the least squares linear fit to the data). The data are very well fitted to a second order

polynomial (solid line with R squared equal to 0.99974) which, in turn, serves as a precise means of evaluating energy response based on the thermistor response. The data of Fig. 4 show that the response is linear to within the precision of the measurements (R squared equals 0.999931). We have opted to use theunjacketed cells since the response is more than adequate and we minimize the alteration of other temperature related phenomena (i.e. deuterium solubility in palladium).

In operation, the thermistor data are measured and recorded by the Asyst(TM) program. Simultaneously the data are transformed into heat units and plotted on the computer screen as the output power from the cells. A simultaneous plot of the input power ( $E \cdot I$ ) is plotted and recorded for each cell.

b. A second calibration procedure is performed at any time during the electrochemical process as long as the input power is constant over the period of time required to carry out the calibration. This is a relatively rapid process for unjacketed cells, requiring ca. 5-10 minutes per point as depicted in Fig. 5. This process is identical to the "standard addition" analyses, with back extrapolation to "zero" (the bath temperature in this instance). Although thermistors are not used for absolute temperature measurements, it is worthy of note that the millivolt readout value corresponding to the bath temperature did not change over the weeks of monitoring the electrochemical process that was responsible for the heat output.

In operation, the direct measurement process is used continuously to account for all heat regardless of variations encountered (current changes, electrolyte replenishment, bath filling, etc.), and calibration checks are used periodically for verification.

### C. HEAT SOURCE ANALYSES

The rate of heat generation,  $W$ , within an electrochemical cell can be viewed as a separate source. Each source is tabulated and a discussion of the evaluation methodology is presented. In view of the dynamic character of these calorimetric/electrochemical analyses we will reserve the classical designation,  $Q$  (i.e. joules, calories, British thermal units, etc.), for the integral measure of energy and use the  $W$  symbol for the dynamic rate of energy production (i.e. Joules/second, watts, calories/second, etc.).

#### 1. JOULE HEAT, $W_J$ .

This is the heat evolved as current flows across the resistive medium (aqueous electrolyte) between the anode and cathode.

Evaluation is straightforward for the case where both the anode and cathode are composed of identical pure metals:

$$W_J = VI$$

where  $V$  is the electrical potential (voltage) between the anode and the cathode and  $I$  is the current (amperes) passing under the imposed voltage. However, in the present instance these electrodes are dissimilar and an electrochemical potential ( $V_0$ ) exists due to this difference in chemical potential and/or difference in concentration of constituents within the electrode matrix. Thus the Joule heat is realized only after the imposed potential exceeds the equilibrium (no current flow) state, leaving us with the correct relationship:

$$W_J = (V - V_0)I \quad (1)$$

(This can be related to every day occurrences such as recharging an automotive storage battery -- no current flow will occur until the imposed voltage exceeds that of the combined cell voltage.) In the strictest sense  $V_0$  should be measured at no current drain (high impedance volt meter) and at zero separation from the anode surface. Standard practice involves the measurement with a high impedance voltmeter at a series of distances from the anode, followed by algebraic extrapolation to the true value at "zero" separation.

An alternative method of analysis can be made if an accurate measure of the resistance between the electrodes is available. In principle one can calculate the resistance of a solution of known composition (known conductivity) and precisely define electrode geometry (size, shape, position, etc.). If an accurate measure of the cell resistance is available, then one can evaluate the Joule heat as follows:

$$W_J = I^2 R(\text{cell}) \quad (2)$$

One must exercise due care and caution so that the resistance measured is just that, and that it does not include an impedance component due to the electrochemical potential mentioned above.

## 2. ELECTROLYTIC DISSOCIATION HEAT, $W_D$ .

When one considers normal water as the solution phase for the electrolytic medium, the reaction is



with well defined enthalpy related to the standard enthalpy of formation of liquid water (25 C and one atmospheric pressure)<sup>2</sup>

$$H(\text{diss}) = - H^\circ(\text{H}_2\text{O}) = 285838 \text{ J/mol.} \quad (4)$$

One can calculate the rate of electrolysis of water as  $I/F$  (moles/sec), where  $F$  is the classical Faraday constant (coulombs/mole). The dissociation heat is then evaluated as

$$W_D = [I \ H(\text{diss})]/F \quad (5)$$

such that the numerical values are

$$W_D = 2.9625 \ I \text{ (J/sec) \{for normal water\}}$$

This approximation is valid in the region of dilute solutions where the water is for all intents and purposes in the same thermochemical state as that of liquid water. Additional corrections must be made for systems where the temperatures and pressures are appreciably different from those of the standard states. These are straightforward and are defined in standard texts on the subject<sup>3</sup>.

### 3. EVAPORATION HEAT, $W_V$ .

As the hydrogen and oxygen are generated at the surfaces of the electrodes they pass upward through the aqueous electrolyte as very fine bubbles. Water is thus entrained to virtual saturation (100% relative humidity) and we must account for the endothermicity of the process



$$H(\text{VAP}) = 44013 \text{ J/mol} \quad (7)$$

The amount of gas released is given by equation (3) and the amount of entrained water will be related to the vapor pressure of water,  $P(\text{H}_2\text{O})$ , at the temperature of the experiment<sup>4</sup>. The heat effect is readily calculated:

$$W_V = \{1.5 [P(\text{H}_2\text{O})/P(\text{ATM})] [H(\text{VAP})/F]\} I \quad (8)$$

As an example

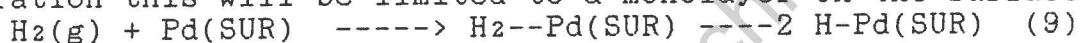
$$W_V = -0.02251 \text{ J/sec \{25 C cell temperature, normal water, at sea level pressure\}.}$$

This relation is valid for dilute solutions, but corrections to the vapor pressure of water over saturated solutions are readily available or its magnitude can be measured if need be. If vigorous release involves large bubbles of hydrogen and/or oxygen, they may not become water saturated and the true relative humidity will be required (a direct multiplicative correction factor in equation (8)). Small corrections for overpressure of

the electrochemical cell, barometric fluctuations, and/or altitude effects are required for the ultimate analytical precision. (The pressure in the laboratory should be measured since most modern facilities operate at pressure that differ from than that of the world outside.) These corrections are of lower order when viewed with respect to the first order effects involved in  $W_D$ .

#### 4. HEAT OF ADSORPTION, $W_A$

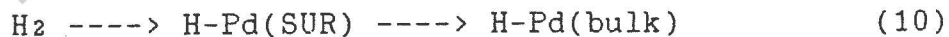
A portion of the hydrogen generated at the anode will be adsorbed onto the surface of the metal. This is a rather rapid process and will be related to the concentration of hydrogen in the aqueous phase. When operating at overpotentials to assure saturation this will be limited to a monolayer on the surface.



The mechanism of adsorption varies in complexity for various metals and for the geometry of the surface. In general the enthalpy of adsorption varies with coverage. Since we are interested in the heat evolution after extended periods of equilibration, the correction due to the heat of adsorption will not be evaluated or applied to the experimental data. Even though there may be a considerable amount of transport into the electrode matrix via the adsorbed state, the amount of material in the adsorbed state remains constant (at least as long as the cell current is held constant).

#### 5. HEAT OF ABSORPTION, $W_I$

Metals such as palladium absorb (imbibe) hydrogen and deuterium into the inner reaches of the matrix where solid solutions and/or interstitial alloys are formed. The rate of absorption is dependent upon the concentration of adsorbed hydrogen (deuterium) on the surface and upon the diffusion rate within the matrix.



The hydrogen in the bulk of the palladium resides on lattice sites tetrahedrally and octahedrally coordinated to the Pd atoms, virtually completely ionized in the form of an interstitial alloy. Enthalpy change associated with reaction (10) is given as

$$H_I = - 39.7 \text{ kJ/mol} \quad (11)$$

at low H concentrations and

$$H_I = -40.94 \text{ kJ/mol} \quad (12)$$

at high concentrations of H in the palladium matrix.

The reservoir for the diffusion controlled process is the adsorbed monolayer which is rapidly replaced in a dynamic system where additional adsorption occurs instantaneously. Thus a diffusional gradient will be established. The diffusion constant,  $D$ , is less for hydrogen than that for deuterium and must be used with the time factor,  $t$ , as apropos for a given setup. This discussion is relevant for the standard interstitial imbibed alpha and beta phases in palladium. If the extensive exposure to the electrochemical potential induces some form of "super charging," then the proper enthalpy term must be evaluated. Each type of reaction is characterized by a specific enthalpy term. For instance, if the lattice parameter is forced beyond the 4.035 Angstroms of the "saturation" level of 0.694 hydrogen per palladium<sup>5</sup>, the expansion will involve an enthalpic term markedly different from equations 12 and 13.

In the extreme, one can consider the ultimate limit of time and heat where the cell current will control the influx of hydrogen into the lattice (all electrolyzed hydrogen passes into the electrode with none dissolving into the aqueous medium nor escaping as hydrogen gas). The minimum time required to fill an electrode of Pd ( $M = 106.4$  g/mol) to its saturation capacity ( $C(\text{sat})$  hydrogen atoms per Pd atom) of

$$t(\text{sat}) = \frac{C(\text{sat}) \cdot m(\text{Pd}) \cdot F}{M \cdot I} \quad \text{seconds} \quad (13)$$

$$t(\text{sat}) = 12,600 \text{ seconds} = 3.5 \text{ hours} \quad \text{for 10 g of Pd at a cell current of 0.5 amps}$$

In an operating cell the time for attaining this saturated state will be appreciably longer. Thus the time required to condition electrodes<sup>1</sup> is seen to be reasonable where diffusional processes and gas evolution decrease the filling efficiency. When one considers supercharging the time span will be lengthened even further.

The absorption process can be modelled and the heat evolved calculated based on known phenomena. However it is likely that there are unknown sorption mechanisms that have not been defined at this writing.

## 6. HEAT OF HYDRIDE FORMATION, $W_H$

In the present context we refer to distinct stoichiometric compounds as hydrides. These are generally salts with an appreciable degree of ionic character (i.e. lithium hydride is an excellent example where the lithium is present as a cation, with almost complete transfer of an electron to form adjacent hydrogen

anions). Some metals form both hydrides and interstitial alloys. Palladium is unique in that it does not form a stoichiometric hydride under the conditions studied previously. That is not to say that such hydrides could not be formed in the "supercharged" state formed under the influence of the electrochemical potential over prolonged periods of time. Hydride compound formation is possible and investigators need to examine all possibilities, even to the point of querying the possibility of formation of lithium compounds and/or alloys. Normally the overpotential is too great for lithium reduction at pure Pd electrodes; however, when the material is fully loaded (or even super loaded the surface may be activated enough to allow lithium reduction at the voltages used for cold fusion studies.

The reader is referred to the standard thermochemical references for the relevant data pertaining to the electrodes of interest. In this manner the appropriate correction term with the proper kinetic factor can be applied to the calorimetric analyses.

#### 7. HEAT OF RECOMBINATION, $W_R$

Transition metal surfaces are well known for their ability to serve as catalysts for the recombination reaction



$$H = -285838 \text{ J/mol} \quad (15)$$

This term is difficult to evaluate from first principles, but it will probably be significant to some degree since the magnitude of the heat effect is so great (thermochemical equivalency to the dissociation reaction of opposite sign). The rate of this reaction will be dependent to a degree on the rate of dissociation but will be somewhat unpredictable based on the known occurrence of autocatalytic and surface poisoning of commercial catalysts. In addition, small amounts of reaction on active surfaces have been known to lead to "hydrogen spillover" where adjacent inert surfaces are made catalytically active. The exothermicity of this reaction could well be attributed to cold fusion processes if not recognized. First analyses would predict that this heat will be emanated at a constant rate as long as the overvoltage is great enough to generate appreciable amounts of gases.

$$W_R = k_R t \quad (16)$$

#### 8. HEAT OF PURGE EVAPORATION, $W_P$

The use of a purge gas such as argon, nitrogen, or helium can serve two purposes. First, the hydrogen and oxygen generated in the dissociation reaction will be carried from the electrochemical cell more quickly and decrease the amount of the exothermic recombination reaction. Second, the temperature homogeneity within the cell can be improved by judicious placement of the purge tube orifice. Purging is not always used, but it surely must be taken into account if employed. Reaction (6) is applicable and will load the purge gas via a mechanism akin to the evaporation process described above

$$W_p = \left[ \frac{P(H_2O)}{P(PUR)} \right] \left[ \frac{H(VAP)}{F} \right] R(PUR) \quad (17)$$

where  $P(PUR)$  and  $R(PUR)$  are the pressure and rate (moles per second) of purge gas introduction into the electrochemical cell, respectively.

$$W_p = 0.03289 R(PUR) \quad \{\text{for normal water at } 25^\circ \text{C, and one atmosphere pressure.}\}$$

## 9. HEAT OF STIRRING, $W_M$

Accurate calorimetric measurements are facilitated by stirring (mixing) the electrolytic medium to assure temperature homogeneity throughout. Hot spots within the calorimetric vessel alter the heat transfer characteristics and can lead to errors and/or spurious results. A trade off is involved to generate a minimum amount of internal temperature variance, balanced against a minimum amount of heat generation by the stirring process itself. Cell design and propeller shape are optimum when a minimum of streamlines are cut in the continuous flow of liquid. Constant speed of stirring is imperative, especially for the long term power measurements proposed for electrochemical fusion studies. Ideally

$$W_M = k_M \quad (18)$$

where  $k_M$  is a constant independent of current and time for a given stirring rate and cell geometry.

## 10. MISCELLANEOUS HEAT EFFECTS, $W$

Several sources of error are inherent in these and virtually all other calorimetric measurements. These are of secondary importance but must be recognized for accurate work:

- a. Absorption of electromagnetic radiation is to be avoided unless a full account can be made. Our cell is surrounded

by a water bath that is transparent to ultraviolet and absorbs virtually all of the infrared radiation before it impinges on the sample chamber. The glass cells are transparent to the ultraviolet. Similar arguments are true for sonic and ultrasonic radiation, although such sources are not normally found in calorimetric systems.

b. When any portion of the cell is at a lower temperature than that of the bath, one must account for the different direct heat leak path. Direct condensation on these surfaces can lead to a "heat pipe" effect where water exothermically condenses on the surfaces and subsequently returns to the electrolyte. This cyclical process can lead to errors in calculating heat effects. We have used polytetrafluoroethylene, with its hydrophobic surfaces, as a construction material in all areas other than the glass contact with the isothermal bath.

c. The dynamic electrochemical systems require that the water must be replenished and the ultimate heat transfer will depend upon the electrolyte level in the cells. Two aids can minimize this effect. Catalytic recombination of the gases in the head space of the cell will diminish the amount of loss by direct gasification and by entrainment. The other method involves continuously replenishing the water, either at a calculated rate or by means of coupled servo control related to the electrolysis current. Ultimately one should be sure that the added water (and electrolyte, when needed) is at precisely the same temperature as the reaction medium. Alternatively, the data may be corrected if the temperature and composition of the additive is known. At this writing we have not expended the time or effort to develop the supplemental continuous addition techniques, since the correction calculation is so simple.

other minor factors are present, but at this writing, none have been noted to contribute to the calorimetric analyses performed in our laboratory.

## 11. NUCLEAR FUSION HEAT EFFECTS

Although the electrochemical studies described here are virtually identical to those of others, we have made no direct observation of any nuclear event in an operating calorimetric cell. The "cold fusion" postulate is often based upon unexplained heat effects which often are explained by default to be of nuclear origin. Suffice it to say that tremendous amounts of energy are released by fusion events and that we will present detailed analyses if and when we note heat in excess of that explained by chemical phenomena.

## D. REFERENCES

1. M. Fleishmann, S. Pons and M. Hawkins, "Electrochemically Induced Nuclear Fusion of Deuterium," J. Electroanalytical Chemistry 261 301 (1989).
2. F.D. Rossini, D.D. Wagman, W.H. Evans, S. Levine, and I. Jaffe, Selected Values of Chemical Thermodynamic Properties, National Bureau of Standards Circular 500, 1952.
3. M.A. Paul "Principles of Thermodynamics" McGraw-Hill, 1951.
4. R.W. Bain, STEAM TABLES: Physical Properties of Water and Steam, Her Majesty's Stationery Office, 1964.
5. H.J. Goldschmidt, INTERSTITIAL ALLOYS, Plenum Press (1967).

## E. FIGURES

Fig. 1. ELECTROCHEMICAL CELL FOR CALORIMETRIC ANALYSES. The essential components are noted schematically; 1. Anode, 2. cathode, 3. Thermistor, 4. Calibration Heater, and 5. Stirrer. Not exactly to scale.

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# ELECTROCHEMICAL CALORIMETER CELL

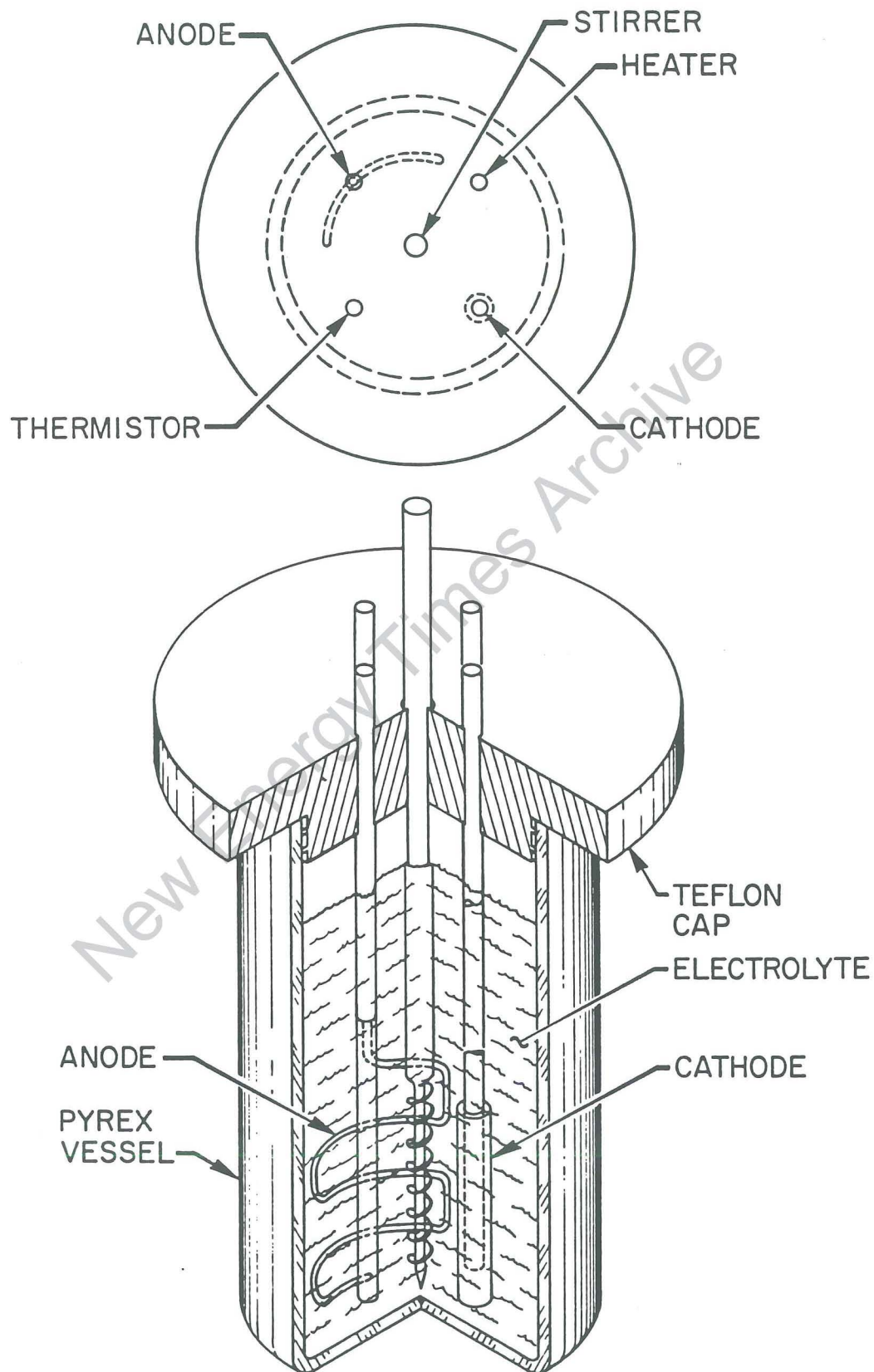


Fig. 2. CALIBRATION DATA FOR WATER-JACKETED CALORIMETER CELL. Data span approximately 4 deg. C and the horizontal axis. The dashed line is seen to give a poor least squares linear fit to the data. The second order fit is quite good as noted in the solid line.

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CELL 2h WITH JACKET: BATH REF

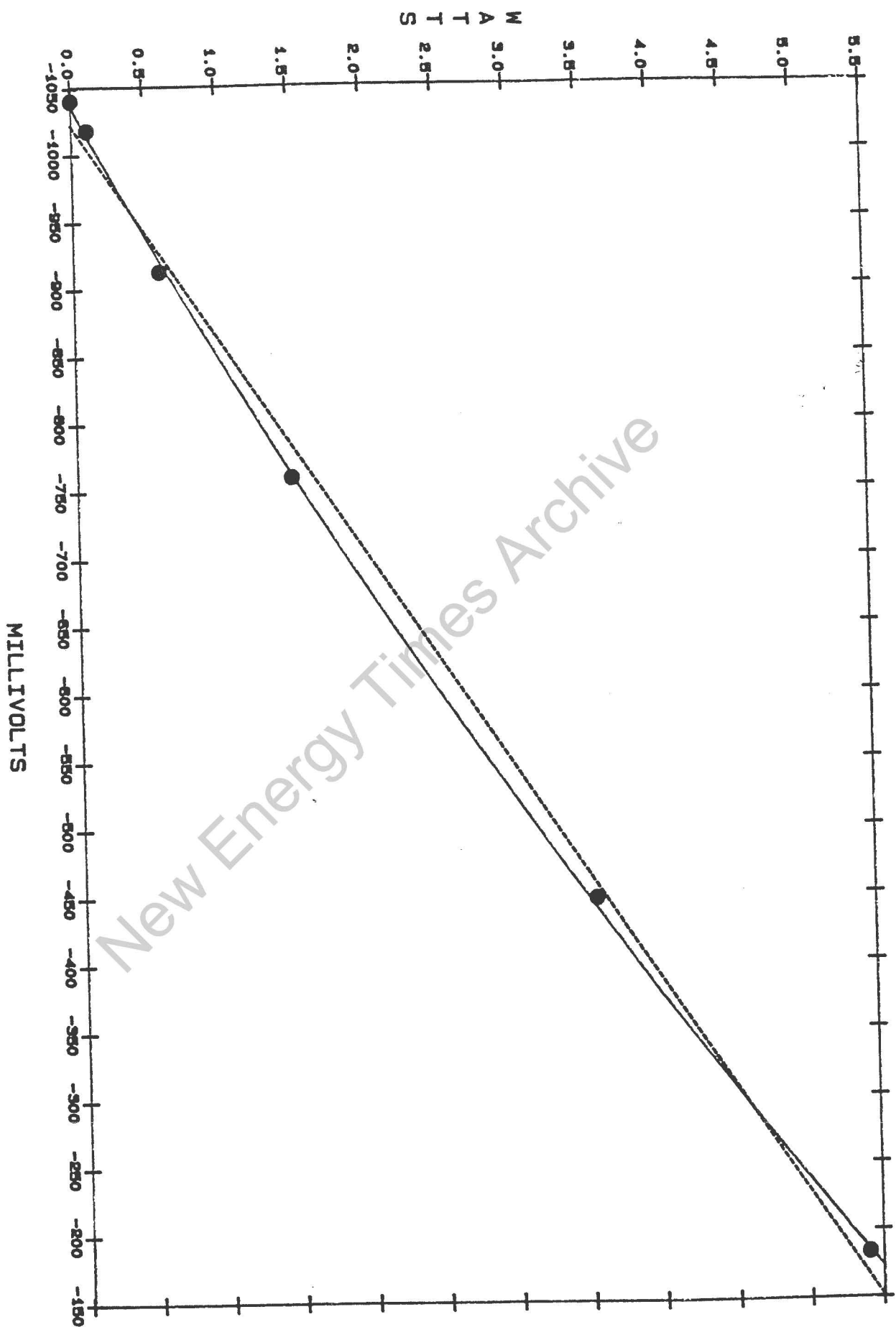


Fig. 3. CALIBRATION DATA FOR CALORIMETER CELL WITHOUT WATER JACKET. The excellent degree of linearity is noted over the approximately 2 deg. C range of the data.

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CELL 2H WITHOUT JACKET

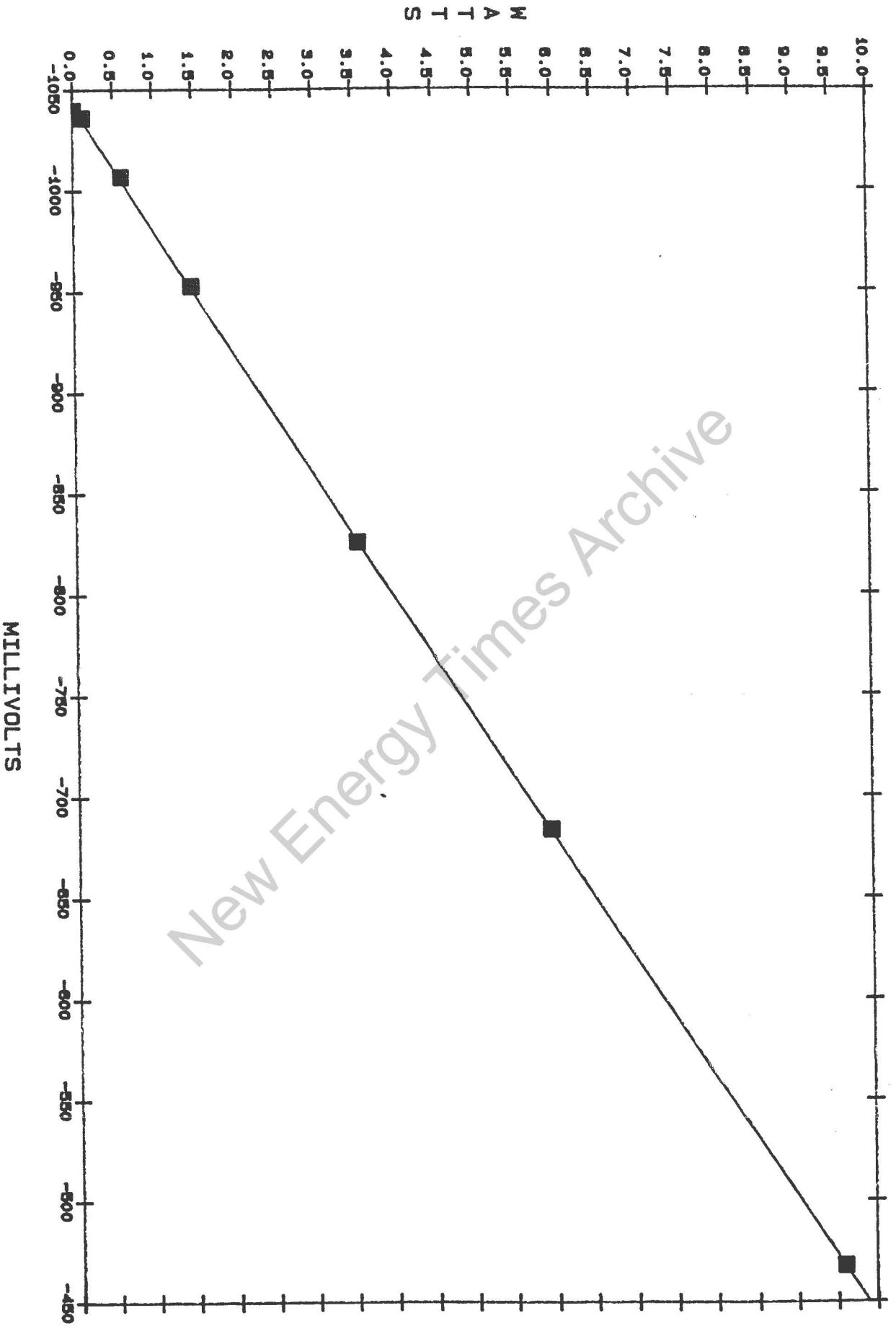
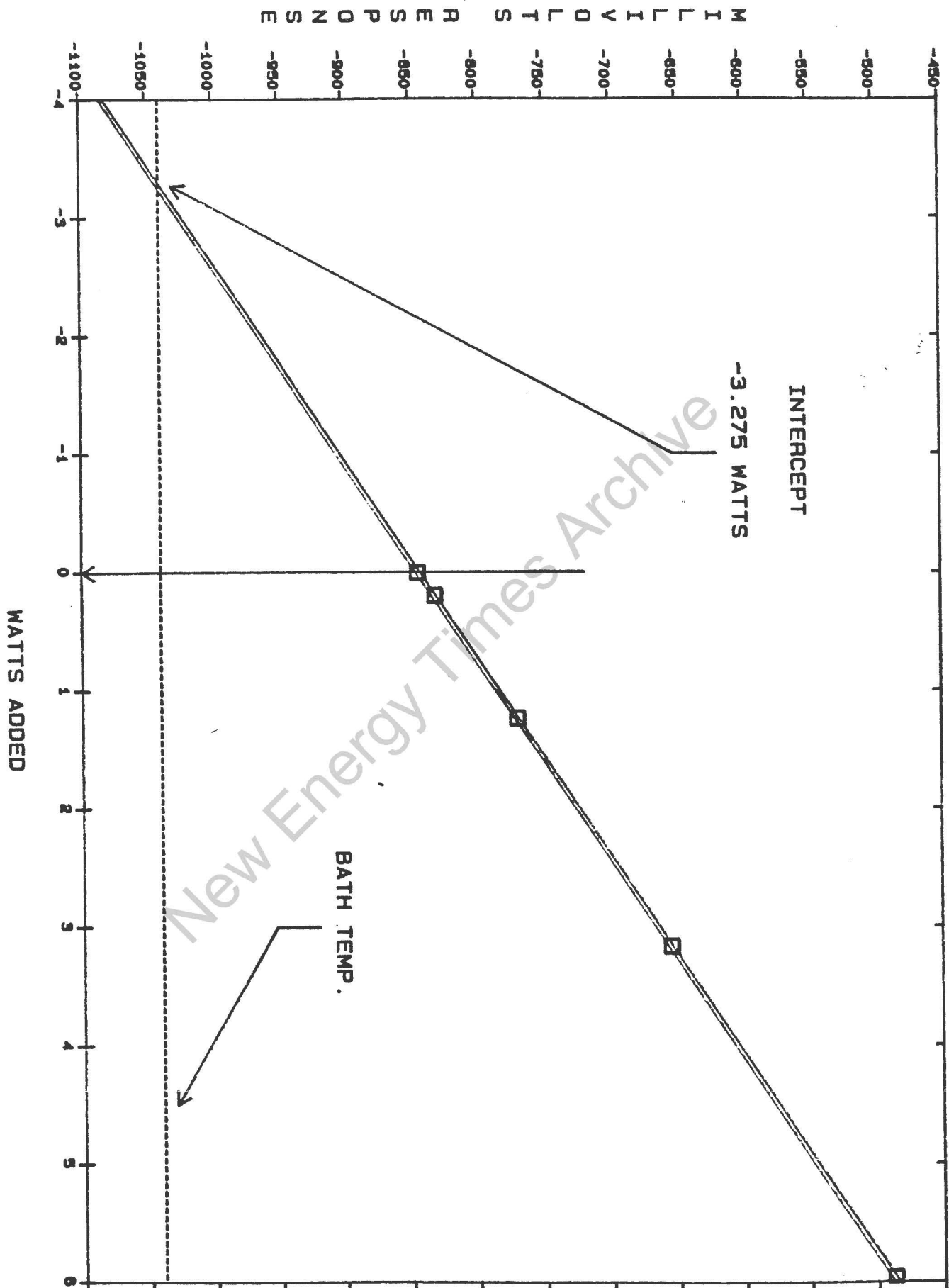


Fig. 4. ONLINE CALIBRATION OF THE ACTIVE CELL WITHOUT THE WATER JACKET. This technique measures the heat input at any time by the additive technique. At any given time the amount of heat is measured by the vertical displacement of the calibration line above the horizontal line with respect to the bath temperature at the horizontal position corresponding to "zero" electrical input.

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### III. NEUTRON COUNTING EXPERIMENTS

#### A. OVERVIEW

The announcement by Pons and Fleischmann in March 1989 that cold fusion had been achieved by using heavy water (deuterium oxide) in electrochemical cells with palladium cathodes and platinum anodes sparked a worldwide effort to duplicate their results. The initial experiments conducted in the Metals and Ceramics Division at Oak Ridge National Laboratory concentrated on detecting neutrons. This was thought to be the quickest and easiest means of proving the existence of fusion of deuterons.

Because of a lack of detailed information on the conditions employed by Pons and Fleischmann, a system was designed that allowed for maximum flexibility in experimental conditions. The goal was to bring cells on-line sequentially while exploring different operating conditions and electrode treatments, and to have the capability to rapidly incorporate crucial design information, as it became available, into new cells. The cells were also designed to serve as electrochemical calorimetry cells for a future experiment (in the planning stages when the neutron counting experiments were beginning).

Eventually, five cells with palladium cathodes and one cell with a titanium cathode were involved in the neutron-counting portion of these cold fusion experiments.

#### B. EXPERIMENTAL

The experiment was designed to accommodate six electrochemical cells spaced near the optimum distance (approximately 5 cm) for thermal neutron detection by a central  $\text{BF}_3$  detector as shown in Fig. 1. The final design included a second  $\text{BF}_3$  detector placed in the water bath approximately 0.3 m from the cells to monitor the background. Separate counting circuitry was used with each cell and consisted of an EG&G ORTEC high voltage power supply (Model 456), a TENNELEC single channel analyzer (Model 2031), a Tennelec linear amplifier (Model TC 205a), and a TENNELEC TC909 power supply. Each counting circuit was optimized for counting thermalized californium-252 neutrons using pulse height discrimination. Fig. 2 shows the multichannel output from the cell  $\text{BF}_3$  with the settings for the upper and lower discriminator levels. The output from both the cell and background  $\text{BF}_3$  counters was monitored continuously by a Stanford Research Systems two-channel gated photon counter (Model SR400) with a counting period of 16.66. The  $\text{BF}_3$  detector monitoring the cells

was approximately 2% efficient for thermalized californium-252 neutrons.

The cathodes for the six electrochemical cells used in the neutron counting experiment were as follows: (1) two cold-cast, swaged 3.1-mm-diam by 5-cm palladium rods, (2) one cold-cast 6.25-mm-diam by 5-cm palladium rod, (3) one 44-g cold-cast palladium "button," (4) one cold-cast 10.9-mm-diam by 3-cm palladium rod, and (5) one cold-cast 6.25-mm-diam by 5-cm titanium rod. The anodes were either platinum or platinum/rhodium wire and the electrolyte was 0.1N LiOD. All cells were operated at current densities ranging from 25 to 300 mA/cm<sup>2</sup>.

### C. RESULTS AND DISCUSSION

During the course of this experiment, four count rate excursions above background were recorded. These events are summarized in Table 1 in the order of occurrence. Of the four excursions, the first appeared to be the result of a rising level of neutrons (the MCA gave a low-noise spectrum similar to the one shown in Fig. 2), but the electrochemical cells were ruled out as the source. The last excursion was similar to the one described above (a slow rise in count rate over 18 hours or more) and was shown by the multichannel analyzer to be the result of an unknown noise source.

Excursions 2 and 3 in Table 1 occurred while the experiment was unattended. A thorough check of the neutron counting system after each event showed everything to be in proper working order. Thus, neutron emission by one of the six cells cannot be ruled out. Excursion 3 is unique among the four events because the apparent rise in neutron count rate occurred in several "bursts" lasting an hour or less, as opposed to the slow rise and fall in count rate observed in the other events. Fig. 3 shows the outputs of both the background and cell BF<sub>3</sub> detectors during this event. The largest peak in Fig. 3 corresponds to 11.7 n/sec (rising from background average of 2.3 n/sec), and is over 27 standard deviations above the background.

The experiment with the large 44-g "button" was terminated when the cell was destroyed by an explosion, presumably D<sub>2</sub> + O<sub>2</sub>. Since this occurrence, the cell and background BF<sub>3</sub> counters have shown no variations above background (for several months), and all attempts to duplicate these results by varying experimental conditions in the remaining cells have been unsuccessful.

Detailed metallurgical and chemical analyses of the 44-g "button" and one 6.25-mm-diam rod have shown that palladium charged with deuterium under electrochemical conditions shows a remarkable ability to retain deuterium. The D/Pd ratio (measured from x-ray lattice parameters, nuclear microanalysis and thermogravimetric

analysis) in electrodes exposed to atmospheric conditions typically has remained above 0.5 over many months. Metallurgical analysis of after-use electrodes has shown a slow transition from the beta-phase, but with no detectable alpha-phase. No evidence of fusion products has been found inside the electrodes.

#### D. REFERENCE

1. E.S. Pons, M. Fleischmann, and M. Hawkins, J. Electroanal. Chem. 261 301 (1989).

TABLE 1. EXCURSIONS IN NEUTRON COUNTING EXPERIMENTS			
1	Slow rise and fall in count rate	~18 h	No background BF <sub>3</sub> . Could not be correlated with cells.
2	Slow rise and fall in count rate	~24 h	Background BF <sub>3</sub> remained flat. Unattended.
3	Two large increases in count rate	45 and 60 min	27 standard deviations above background. Unattended.
4	Slow rise and fall in count rate	> 24 h	Shown to be noise by MCA.

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## F. FIGURES

Fig. 1. NEUTRON MONITORING SYSTEM FOR ELECTROCHEMICAL ANALYSES OF DEUTERIUM IN METALS. A reference detector is used to monitor background effects and a sampling detector monitors up to six cells mounted circumferentially.

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# ELECTROCHEMICAL CALORIMETRY SYSTEM

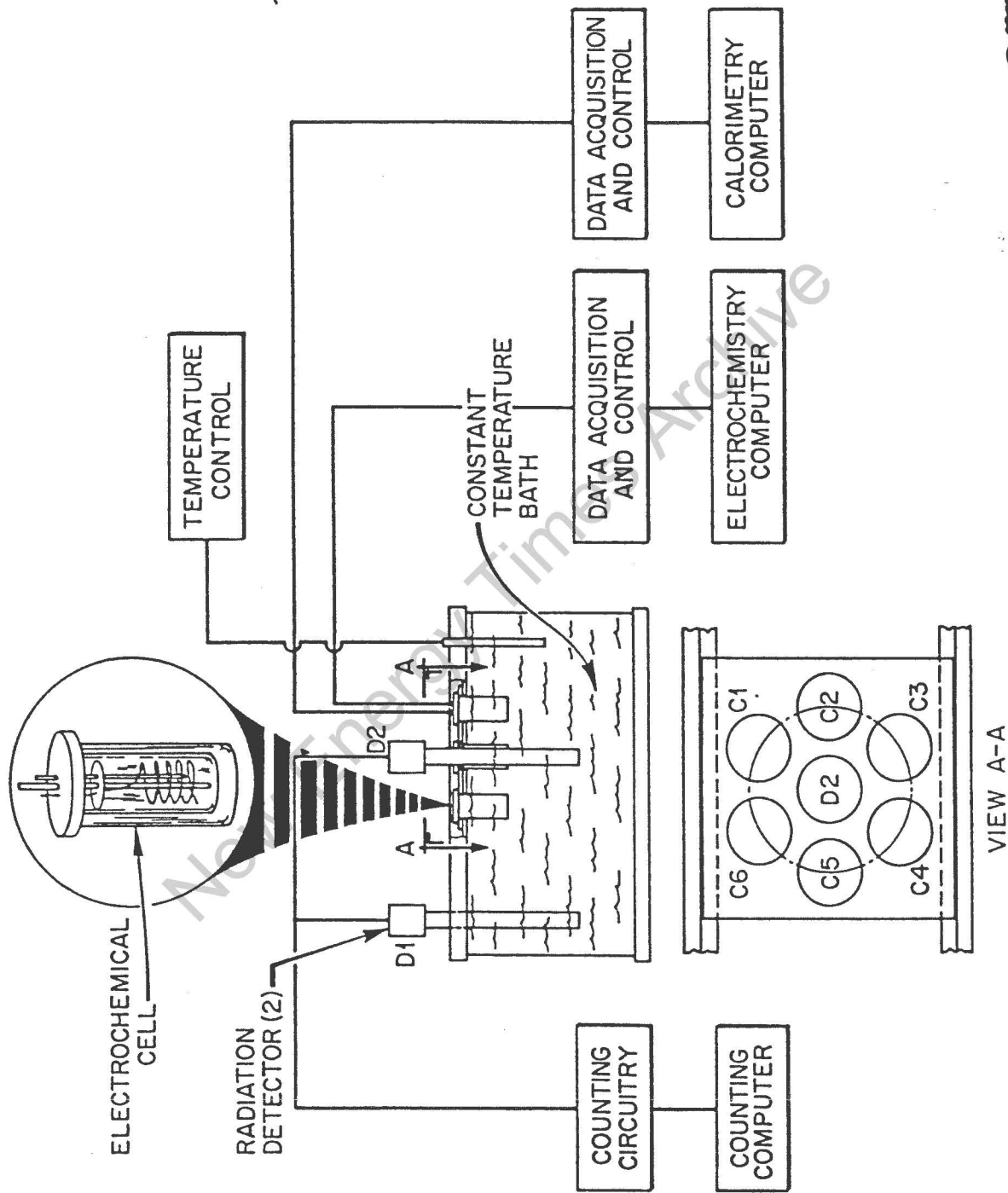


Fig. 2. MULTICHANNEL ANALYSES OUTPUT FOR CUMULATED NEUTRON COUNTS: AUG. 25, 1989. Upper and lower discriminator levels were set to avoid analyzing electronic noise. Unusually large noise input is registered in these traces if and when it is present.

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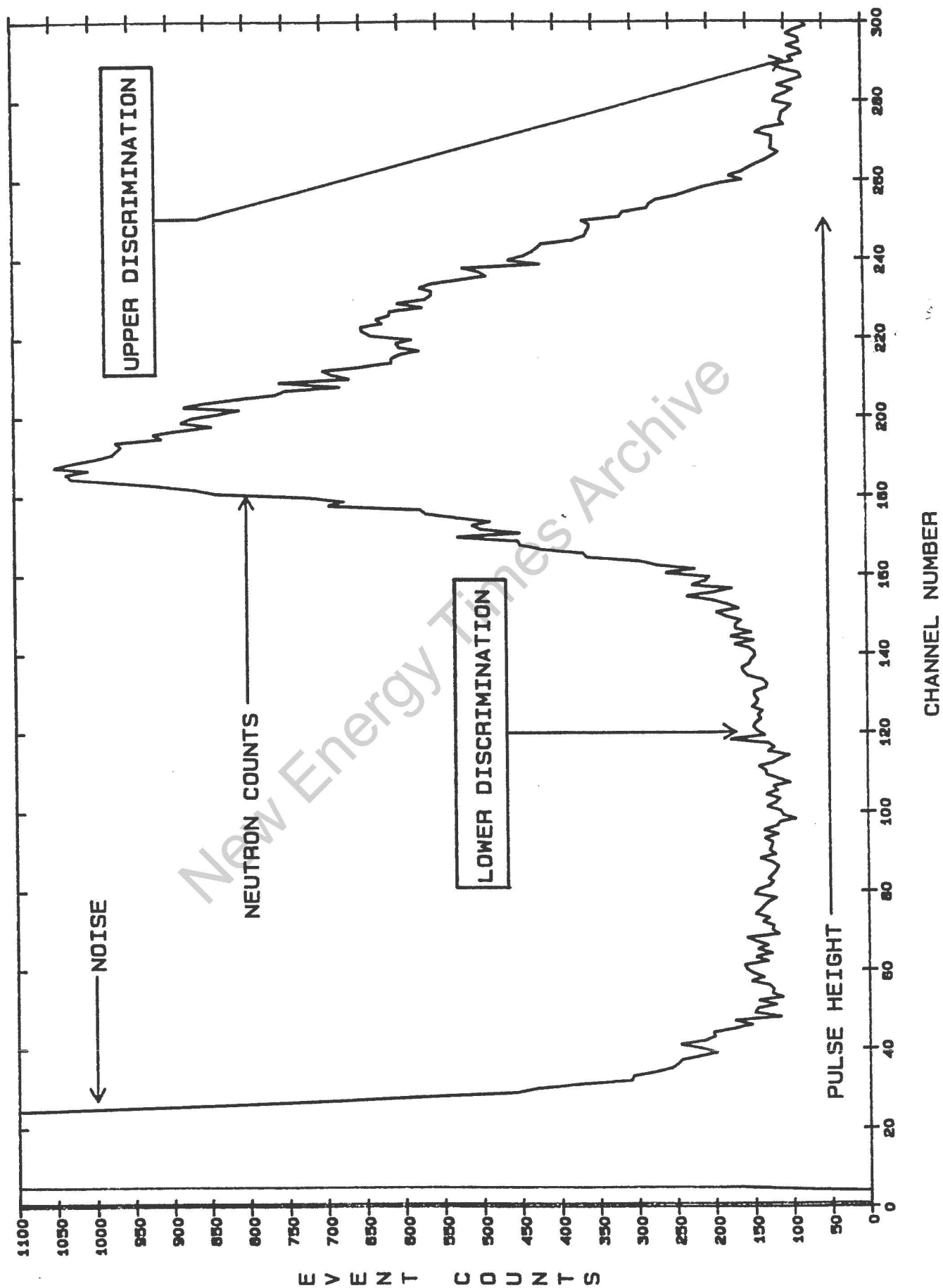
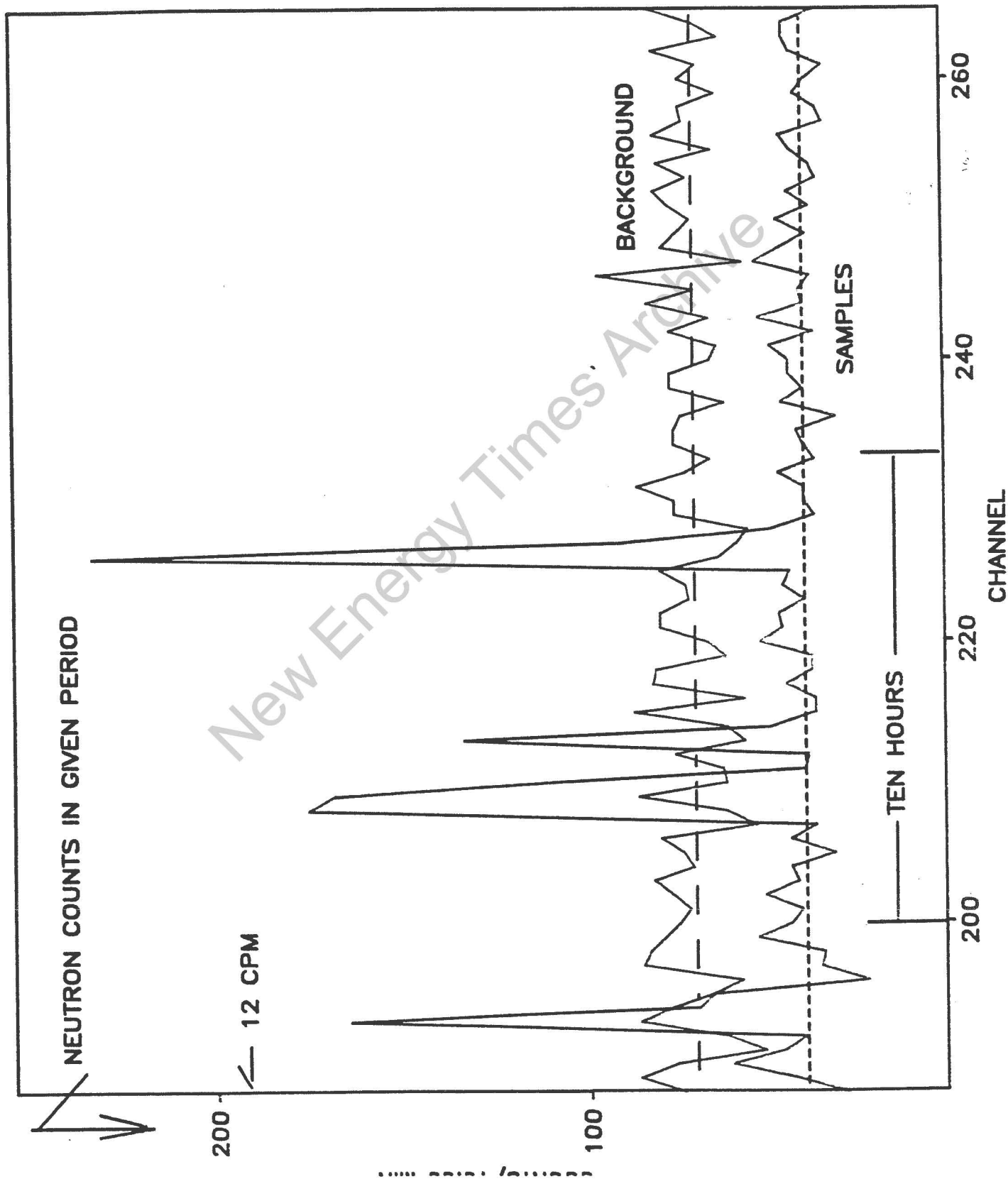


Fig. 3. COUNT RATE EXCURSIONS IN NEUTRON MONITORING SYSTEM. In every case the sampling counter gave positive response while the background counter remained unperturbed. The first excursion (far left) was the result of microphonics as the counter was bumped while placing cells in the holder.

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#### IV. TRITIUM MONITORING ANALYSES

##### A. OVERVIEW

In the event of nuclear fusion of deuterium, tritium production should be observed. Since tritium production has been reported by several researchers, we have begun to monitor our systems for tritium.

##### B. EXPERIMENTAL

Samples of electrolyte are acquired on a routine basis (ca. every 2-3 days). Full replenishing of electrolyte is made using LiOH or LiOD solutions for the respective electrolytic media. These aliquots are sealed in polyethylene vials for transport to the analytical laboratory in the Y-12 facilities. This facility is used routinely for tritium assay and for trace analyses. The scintillation detector is calibrated with standards at the same time that the samples are measured. All cells (both heavy and light water) were monitored for tritium. In every case the electrolyte solutions, replenishing solutions, and solvents were of low tritium content: Aldrich Chemical Co.; 100 DPM/ml. In addition a single cell constructed of yellow glass, equipped with a gold anode, and fitted with a palladium cathode (cold cast, 6 mm diameter and 15 cm length) was placed into operation on JUN 30 1989. The cell volume was ca. 200 cc and was operated at room temperature, 23 +/- 2 °C.

##### C. RESULTS AND DISCUSSION

No increase in tritium content has been detected at this writing for any of the calorimetric cells.

The gold anode of the separate cell began to disintegrate after one week of operation, forming a brown residue. Tritium analyses data are presented in Table 1. The gold anode was completely gone by day 40 and no electrolytic current was passing through the cell. The data are recast graphically in Fig. 1. to show that there was a slow gradual buildup of tritium in the electrolytic medium for ca. 20 days, probably due to the isotopic enrichment factor for hydrogen isotopes in palladium. On day 24 there was an additional enhancement which became very pronounced on day 26. The graphical data of Fig. 1. is very conservative since it uses the average value of the counts. The value of 700+ counts is probably a much more valid estimate of the tritium activity since the counting technique per se cannot enhance the counts. There is a high probability that the basic medium of the low count sample (250+ DPM/ml) may not have been entirely neutralized and the scintillation "cocktail" activity may have been adversely quenched. Direct observation of the cap on the

cell with 250 counts showed some deterioration. Regardless of the weighting procedure, the data show that the tritium activity was appreciably enhanced and that the activity was subsequently diminished by the normal activity of the electrolytic processes.

After the measurement of the high tritium activity, the concentration decrease back to the level consistent with the linear increase due to normal isotope enrichment. A tacit explanatory model for the reaction mechanism must involve a weakly bound state in the aqueous medium which is rather quickly depleted by the normal purging action of the oxygen and deuterium gases. This would indicate that the activity was associated with surface reactions at the palladium electrode. This process apparently generates DT molecules that tend to remain in the solution for a short period of time. The normal purging action of the D<sub>2</sub> and O<sub>2</sub> produced electrolytically depletes the tritium activity after the transitory tritium production process is completed. The possibility of contamination of the electrolyte is nil. No source of tritiated gases exists in the laboratory and if tritiated water species were introduced, the concentration would remain high-- undiminished by gas purging. Unfortunately we were unable to continue the experiment to search for other tritium production events due to the disintegration of the gold anode.

Calorimeter cells (both light and heavy water) are still in operation and are being monitored for tritium activity.

TABLE 1. TRITIUM SCINTILLATION ANALYSES OF ELECTROCHEMICAL CELL. Yellow glass cell equipped with a gold anode and palladium rod cathode. Duplicate aliquots measured for each sampling.

DATE 1989	TIME HOUR	SAMPLE A (DISINTEGRATION/min/ml)	SAMPLE B
JUN 30	1131	96.58	108.57
JUL 3	1200	116.83	116.74
JUL 5	0802	129.76	117.12
JUL 7	1310	128.49	115.90
JUL 10		115.08	122.31
JUL 12	1145	114.21	129.38
JUL 14	1151	128.34	115.68
JUL 17	1023	136.05	131.30
JUL 20	0910	124.83	117.51
JUL 24	1345	168.42	197.97
JUL 26	1344	250.32	717.56
DUPLICATE		252.72	753.85
AUG 4	1054	153.01	148.07

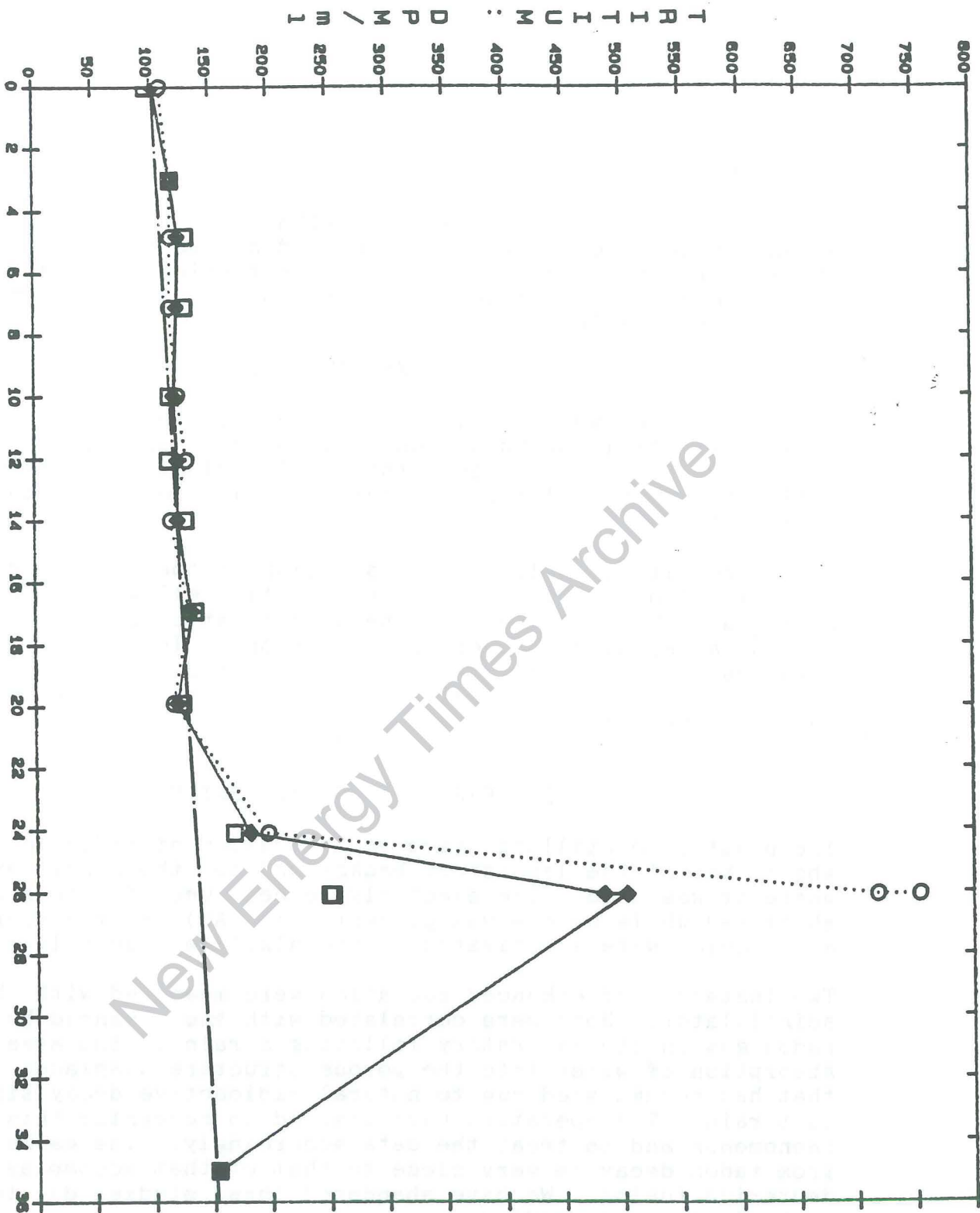
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E. FIGURE

Fig. 1 TRITIUM ENHANCEMENT ACCOMPANYING THE ELECTROLYTIC CHARGING OF DEUTERIUM INTO PALLADIUM CATHODE.

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# DAYS OF ELECTROCHARGING



□ COUNT A  
 ○ COUNT B  
 — AVERAGE  
 --- LIN CONC

## V. GAMMA MONITORING

### A. OVERVIEW

As an aid to discerning the amount and nature of the radiation emitted from the electrolytic cells, screening studies were carried out to attempt to measure radiation emitted from active electrolytic cells.

### B. EXPERIMENTAL

A survey system was used to count scintillation events in a plastic scintillator using conventional counting electronics. The detector was placed in a thin walled aluminum container. Calibration with californium source should the system to be 7 % efficient.

A very sensitive analysis used a sodium iodide single crystal scintillation detector. The electrolytic cell was immersed in a water bath which fitted over the scintillation crystal with a 5 cm annular spacing to thermalize neutrons. This instrument responds to both gamma rays and neutrons and we did not incorporate any electron discrimination to distinguish between the two types of radiation.

### D. RESULTS AND DISCUSSION

The plastic scintillator gave no indication of radiation over and above that of the laboratory background for the single experiment where it was used. The electrolytic cell used for this study was shattered while no one was present (3:30 AM) and the operating electronics were inactivated by the alkaline electrolyte.

Two instances of enhanced radiation were measured with the NaI scintillator. Both were correlated with the enhanced buildup of radon gas in the laboratory following a rain in the area. Absorption of water into the porous structure displaces the radon that has accumulated due to natural radioactive decay since the last rain. The operators have learned to recognize this phenomenon and to treat the data accordingly. The gamma energy from radon decay is very close to that of that accompanying deuterium fusion. We have abandoned these studies due to the lack of time and funding.

## VI. ELECTRODE CHARACTERIZATION

### A. OVERVIEW

At this writing, there are innumerable procedures and "recipes" for producing electrodes that will provide the desired activity (excess heat, neutrons, tritium, etc.). No uniform consensus exists. Our program is designed to draw upon the expertise and experience of the Metals and Ceramics Division to prepare metal electrodes of varying degree of purity and/or crystallinity. Our initial efforts utilized cold cast palladium electrodes. Later efforts used hot cast palladium, annealed palladium, cold worked palladium, and palladium purchased from Johnson-Mathey (ostensibly the same material that the Utah school used in their studies). Separate portions of all electrode material are archived for analytical comparison of the electrodes after the electrochemical reactions.

A large repertoire of analytical techniques is also valuable in evaluating the nature of the loaded electrodes and for monitoring the kinetics and mechanisms of loss of hydrogen/deuterium from the loaded metals. Much is known about the interaction of hydrogen with metals<sup>1-8</sup>. However, many of the details have yet to be elucidated. For instance, the very process of imbibing of deuterium into palladium leads to a 15+ volume percent expansion, changing the chemistry and structure appreciably in the process.

### B. THERMOCHEMICAL ANALYSES

Thermogravimetric Analyses (TGA) and Differential Thermoanalyses (DTA) are straight forward methods for evaluating the chemistry and structure of the electrodes used in these studies. In our normal mode of operation the analyses are performed in a helium gas flow (75 cc/min), using a sample weighing ca. 200 mg and a reference of approximately the same weight. A quadrupole mass spectrometer is used to analyze the gas phase for abundance and speciation. A typical result is shown in Fig. 1. for a sample of a palladium electrode that had been subjected to extensive cathodic electrolysis in heavy water and subsequently exposed to laboratory air for ca. 33 days. At a temperature ramp rate of 10 °C/min, virtually no weight loss is noted below 150 °C, whence the imbibed deuterium is released quantitatively by 300 °C. Virtually no additional material is lost at higher temperatures. The 1.04 percent weight loss is equivalent to 0.555 D/Pd for the natural isotopic abundance of palladium.

Fig. 2 is a presentation of the differential thermal analysis of

the melting process that occurs at ca. 1570 °C. In addition the thermal effects of the deuterium loss and defect annealing are noted at 229 °C and 815 °C, respectively in Fig. 3. Combining the data of these two figures, one can evaluate the relevant parameters shown in Table 1. The methodology used here is based on the assumption that the integrated area under the DTA curves is proportional to the heat absorbed (negative temperature effect in all cases) and that a first order calibration is afforded by the fusion of the annealed palladium at 1570 °C. Assuming the direct proportionality, the quoted values for the heat of annealing and the heat of deuterium desorption are calculated. The value for the latter is in excellent agreement with the data tabulated by Goldschmidt for hydrogen: 39.7 and 40.9 kJ/mole for low levels and high levels of loading, respectively.

For assurance that the phenomena observed here are associated with the electrochemical processes, we have acquired data for a section of the electrode that had not been subjected to the electrolytic processes (a segment of the cold cast material reserved for just such purpose). The data are presented in Fig. 4, where the weight loss is considered to be negligible--indicating the absence of any preloaded volatile component and assuring that the material lost from the deuterated palladium is solely deuterium. The endotherm for the melting process is quite evident and the endotherm for deuterium loss is absolutely undetectable. The annealing endotherm is not noted. In its place is an exotherm which may be associated with an entirely different kind of defect associated with the cold casting process per se.

It is worthy of note that the deuterium level is as high as 0.555 D/Pd after the prolonged period of time following the electrochemical loading. Any simple process of lattice swelling and relaxation for the deuteration process should not involve thermodynamic hysteresis of the magnitude that would lead to so much retention for such a long period of time. Considerably more work is required to define the kinetics and mechanisms of the processes that occur when hydrogen isotopes are taken up electrochemically by palladium and other metals. The melting endotherm is virtually identical to that observed on the electrocharged sample as noted in Fig. 5. The character of the annealing exotherm, shown in Fig. 6, contrasts markedly to that of the electrocharged material. Details of the melting endotherm for the annealed sample are shown in Fig. 7. Annealing by either method leaves an identical material, as far as can be discerned by thermal analyses.

For comparison, Fig. 8 is the results obtained for a well annealed palladium sample when subjected to the thermochemical analyses. Differential thermal analysis shows only the melting endotherm. The results show that both types of annealing peaks described above are free from instrumentation artifacts. To

complete the picture, the melting endotherm for the annealed uncharged material is presented in Fig. 9. This shows the reproducibility of the measurements, and that the final state of the palladium is the same whether the annealing is accomplished a. initially prior to any electrochemical loading, b. induced directly on uncharged palladium in the thermoanalytic instrument, or c. induced in situ subsequent to dehydrogenation in the thermoanalytical instrument.

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TABLE 1. DIFFERENTIAL THERMAL ANALYSES OF DEUTERIUM LOADED PALLADIUM CATHODE. All values expressed in degrees Centigrade.

PEAK NUMBER	ONE	TWO	THREE
LOWER BOUNDARY	137.3	696.3	1536.8
UPPER BOUNDARY	344.4	984.4	1578.2
PEAK TEMPERATURE	229.4	815.2	1568.7
ONSET TEMPERATURE	169.7	706.2	1557.6
OUTSET TEMPERATURE	328.3	947.9	1573.2
PEAK AREA	-59.77	-62.77	-28.54

ENTHALPY			
(kJ/mole Pd)	36.10	37.91	17.24
(kcal/mole Pd)	8.63	9.06	4.12
(kJ/mole D <sub>2</sub> )	40.07		
(kcal/mole D <sub>2</sub> )	9.58		

## C. CHEMICAL ANALYSES

Several methods of analyses of the chemical composition of our electrodes were employed. Table 1. lists the elemental analyses of a typical palladium electrode by spark source mass spectroscopy as performed by the staff members of the Y-12 Plant Laboratory Mass Spectroscopy Group. This specific electrode was cold cast in the ORNL Metals and Ceramics Division facilities. The surface of the electrocharged segment was milled away and a measure of the chemical composition of the bulk of the electrode substrate was thus obtained. The surface associated copper and platinum are picked up by the electrochemical action on the platinum anode and sundry electric supports in the cell. No lithium entities were found to be transported into or onto the palladium electrode, within the sensitivity (ca. 1 wppm).

Gas phase samplings of out operating heavy water cells were analyzed for tritium by the same group at the Y-12 Plant Laboratory. There was no detectable tritium in the gas samples above the ca. 10 ppmv sensitivity of the instruments. Two different groups analyzed portions of out electrocharged palladium cathodes for entrapped helium. Again the results were negative: no excess helium (3 or 4) over and above that found in the control samples. The total sensitivity was quoted to be ca.  $10^{-9}$  moles of helium.

Table 1. CHEMICAL ANALYSES OF COLD CAST PALLADIUM CATHODE (6.5 mm DIAMETER. Values are quoted as parts per million on a weight basis).

ELEMENT	UNCHARGED CONTROL	CHARGED WITH DEUTERIUM	
		BEFORE SURFACE REMOVAL	AFTER SURFACE REMOVAL
Ag		20	
Au		130	
B	2	70	2
Bi		90	
Ca	<1	100	<1
Co		3	
Cu		900	
Fe	2	180	4
Ir		100	
K		40	
Na	<1		<1
Ni	8	40	3
Pb		70	
Pt	45	1700	100
Rh		40	
Si	10	200	7
Sr		15	
Zn		25	

#### D. PHYSICAL AND STRUCTURAL PROPERTIES

An accurate measure of the amount of deuterium held within the metal is afforded by X-ray diffraction measurements<sup>9</sup> as they are related to the swelling of the matrix<sup>10</sup>. The deuterium concentration has also been verified by nuclear microanalysis studies<sup>11</sup>. This method is specific for deuterium and proves that the lattice is indeed maintained in the expanded state by imbibed deuterium. Conceivably, the deuterium could have escaped and the lattice remained open if an elastic limit had been exceeded. Had there been appreciable exchange with atmospheric hydrogen, the nuclear microanalyses would have measured a lesser amount of deuterium. These studies were carried out over an extended period of time to permit evaluation of kinetics and mechanisms of deuterium release from a highly charged palladium lattice. As the deuterium was slowly released the rod became harder (2-3 fold as measured on a diamond hardness scale) and a definite grain structure began to develop as shown in Fig. 10. The original hardness after first removal from the electrocharging system was ca. 46 to 50 DPN and rather uniform over the surface of the electrode material. After 22 days of exposure to the laboratory air the hardness ranged from 86 to 150 DPN with a wide variation over the surface of the palladium metal.

The etched surfaces after partial loss of the deuterium show the spatially periodic structures characteristic of spinodal decomposition processes<sup>12</sup>. In this instance we are concerned with the decomposition of the  $\beta$  phase, known to exist at concentrations of  $D/Pd > ca. 0.5$ <sup>13</sup>. Shirber and Morosin<sup>9</sup> have evaluated the relation between the lattice parameter,  $a$ , and the  $D/Pd$  ratio,  $X$  to be

$$a = 3.913 + 0.180 X$$

for pressure-loaded (5 kbars) samples in the range  $0.6 < X < 0.97$ . Although the lattice constants may vary for a given mode of preparation (ie. avoiding the  $\alpha$  phase by hydriding at temperatures above the 300 C miscibility temperature<sup>10</sup>), the general trends described below are assuredly valid and informative.

In the process of decomposition, regions of the  $\beta$  phase dissociate. Although there are regions almost completely decomposed (see Figure 2), each with its own dimension,  $z(i)$ , we can view the total process as a single dimension,  $Z = \text{sum of all } z(i)\text{'s}$ . Overall then the rate of extension of  $Z$  is retarded as the decomposition proceeds:

$$\frac{dZ}{dt} \propto \frac{1}{Z}$$

or

$$Z \propto t^{1/2}.$$

where  $t$  is the elapsed time since the imbibed deuterium was first allowed to escape unencumbered to the atmosphere. Since the degree of decomposition is directly related to the concentration of deuterium in the sample:

$$Z \propto \{X(t=0) - X(t)\}$$

which leads to the relationship

$$X(t) = X(t=0) - k t^{1/2}.$$

Fig. 11. graphically shows the corresponding correlation of our data. Numerical evaluation of the adjustable parameters allows us to evaluate the relevant quantities and their standard deviations:

$$X(t=0) = 0.803 (0.009) \text{ deuterium/palladium}$$

$$k = 0.00747 (0.00051) \text{ hours}^{-1/2}$$

$$r^2 = 0.9986$$

$$\text{Standard error of estimate} = 0.0081 \text{ deuterium/palladium}$$

The data presented here is not complete enough at this writing to distinguish between two plausible mechanisms. First is an overall loss of deuterium from the bulk of the palladium matrix where each of the microscopic domains are depleted. Second, the phenomena may be more macroscopic where a concentration gradient is slowly established within the rod. The x-ray diffraction and nuclear microprobe both measure the effects in the outer surface region. If the second mechanism is in play the overall loss of deuterium is much slower than that noted in Figure 9. Further work is underway to more completely define the mechanisms of deuterium loss.

The initial hardening of the palladium by the lattice expansion due to the strain (ca. 15%) induced as the interstitial sites are occupied by deuterons is relatively small. The major hardening is due to the interpenetration (intergranular precipitation) of the  $\beta$  phase and its decomposition product. This product is not well defined  $\alpha$  phase material since we were not able to detect the x-ray diffraction pattern corresponding to the identical crystal habit with smaller lattice spacing ( $a(0)$  values). This is undoubtedly related to the hysteresis phenomena observed in the hydrogen/palladium system<sup>14</sup>. Our work is consistent with the work of others where it is noted that, "It is suggested that two processes occur during a hysteresis scan. In

the nearly linear, reversible regions H<sub>2</sub> can be added or removed from both phases without the formation of any additional amounts of either phase. In the regions of the scans which show marked curvature an additional component enters corresponding to the formation or decomposition of the  $\beta$ -phase."<sup>15</sup> The hydriding/dehydriding processes produce copious numbers of dislocations which are known to be favorable sites for interaction with hydrogen (deuterium)<sup>15-17</sup>.

Regardless of the fate of electrochemical fusion, there is a dire need for fundamental information related to the interaction of hydrogen isotopes with palladium, especially at the high loadings encountered in prolonged electrochemical exposure.

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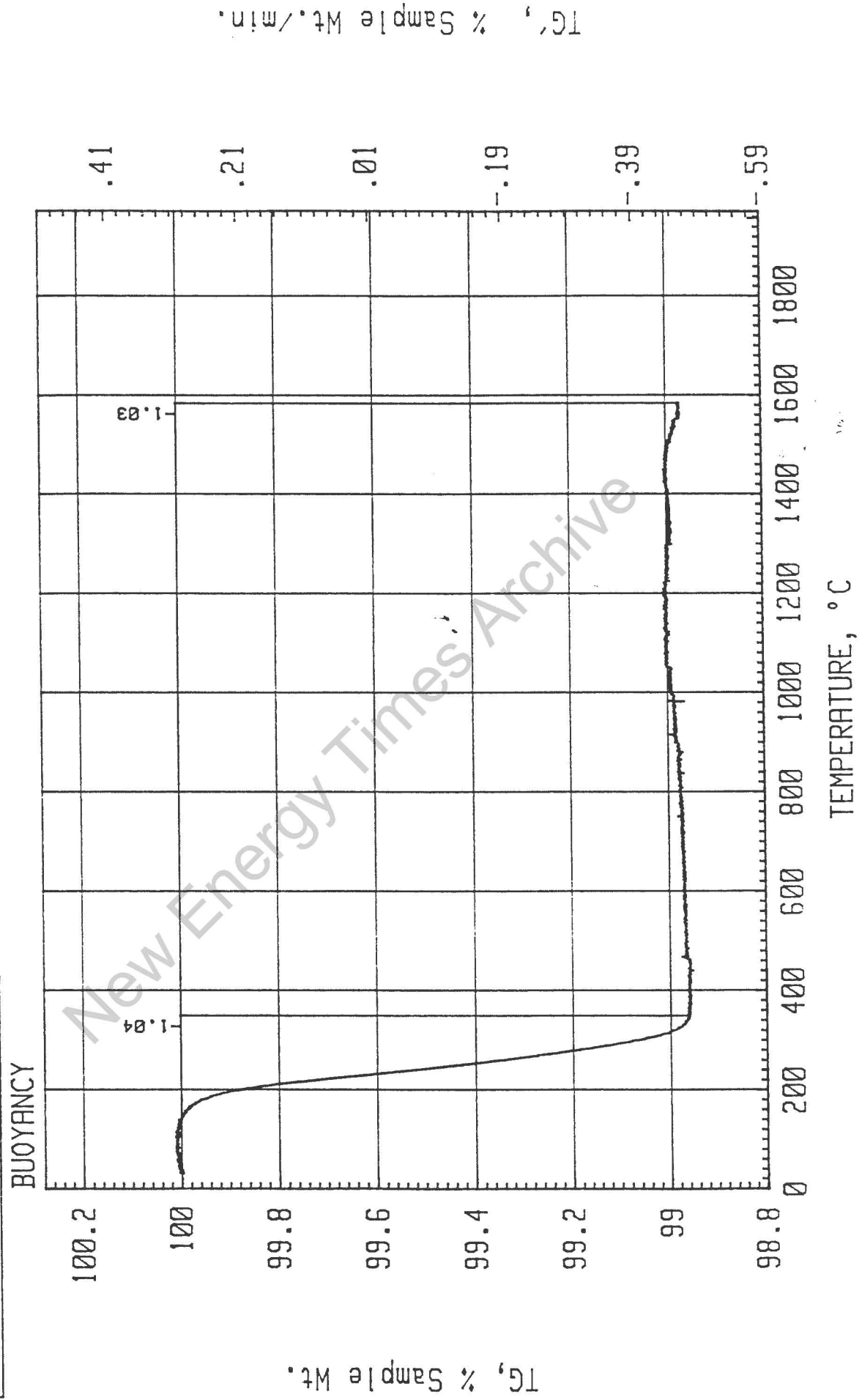
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## F. FIGURES

Fig. 1. THERMOGRAVIMETRIC ANALYSIS OF COLD CAST PALLADIUM  
CATHODE ELECTROCHARGED WITH DEUTERIUM: AFTER PROLONGED EXPOSURE  
TO AIR.

New Energy Times Archive

OPERATOR: WMS		** NETZSCH STA ANALYSIS **		ATMOSPHERE		HE
DATE: 2 Aug 1989		REFERENCE: AL203		FLOW RATE, cc/min:		75
RUN No. 1		CRUCIBLE: AL203		HEAT RATE, °C/min:		10
SAMPLE No. 1		FILE NAME: BI:PD		SAMPLE Wt., mg:		181.4
SAMPLE NAME: PD ELECT		LOT No. 1		REFERENCE Wt., mg:		192.3



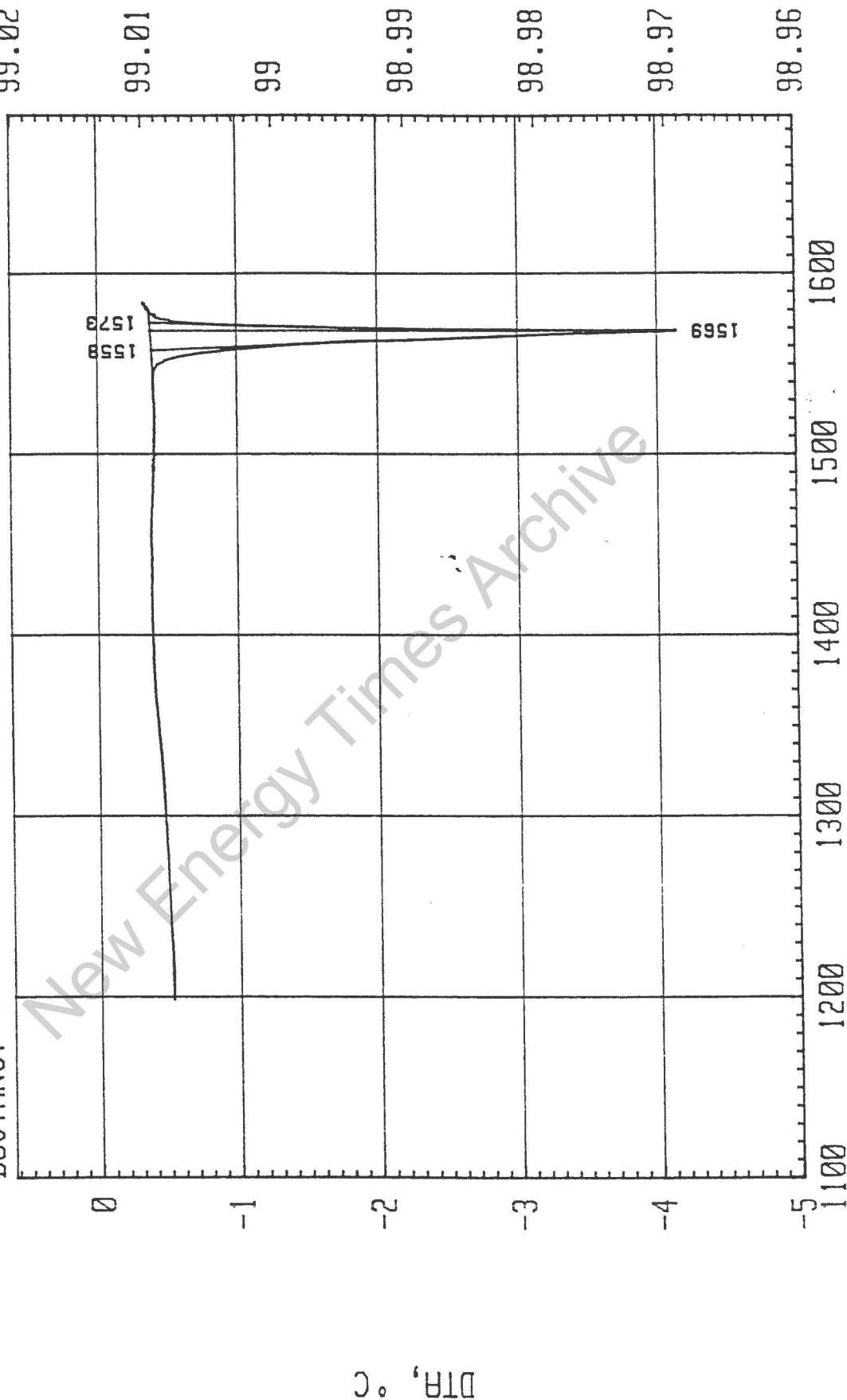
TG, % Sample Wt./min.

Fig. 2. DIFFERENTIAL THERMAL ANALYSES OF THE MELTING OF COLD CAST PALLADIUM CATHODE ELECTROCHARGED WITH DEUTERIUM: AFTER PROLONGED EXPOSURE TO AIR.

New Energy Times Archive

OPERATOR:	HMS	** NETZSCH STA ANALYSIS **		ATMOSPHERE	HE
DATE:	2 Aug 1989	REFERENCE:	AL203	FLOW RATE, cc/min:	75
RUN No.	1	CRUCIBLE:	AL203	HEAT RATE, °C/min:	10
SAMPLE No.	1	FILE NAME:	B1:PD	SAMPLE Wt., mg:	181.4
SAMPLE NAME:	PD ELECT	LOT No.	1	REFERENCE Wt., mg:	192.3

# BUOYANCY



TG, % Sample Wt.

Fig. 3. DIFFERENTIAL THERMAL ANALYSES OF THE LOSS OF DEUTERIUM AND ANNEALING OF COLD CAST PALLADIUM CATHODE ELECTROCHARGED WITH DEUTERIUM: AFTER PROLONGED EXPOSURE TO AIR.

New Energy Times Archive

OPERATOR:		HMS		** NETZSCH STA ANALYSIS **		ATMOSPHERE		HE	
DATE:		2 Aug 1989		REFERENCE: AL203		FLOW RATE, cc/min:		75	
RUN No.		1		CRUCIBLE: AL203		HEAT RATE, °C/min:		10	
SAMPLE No.		1		FILE NAME: B1:PD		SAMPLE Wt., mg:		181.4	
SAMPLE NAME:		PD ELECT		LOT No. 1		REFERENCE Wt., mg:		192.3	

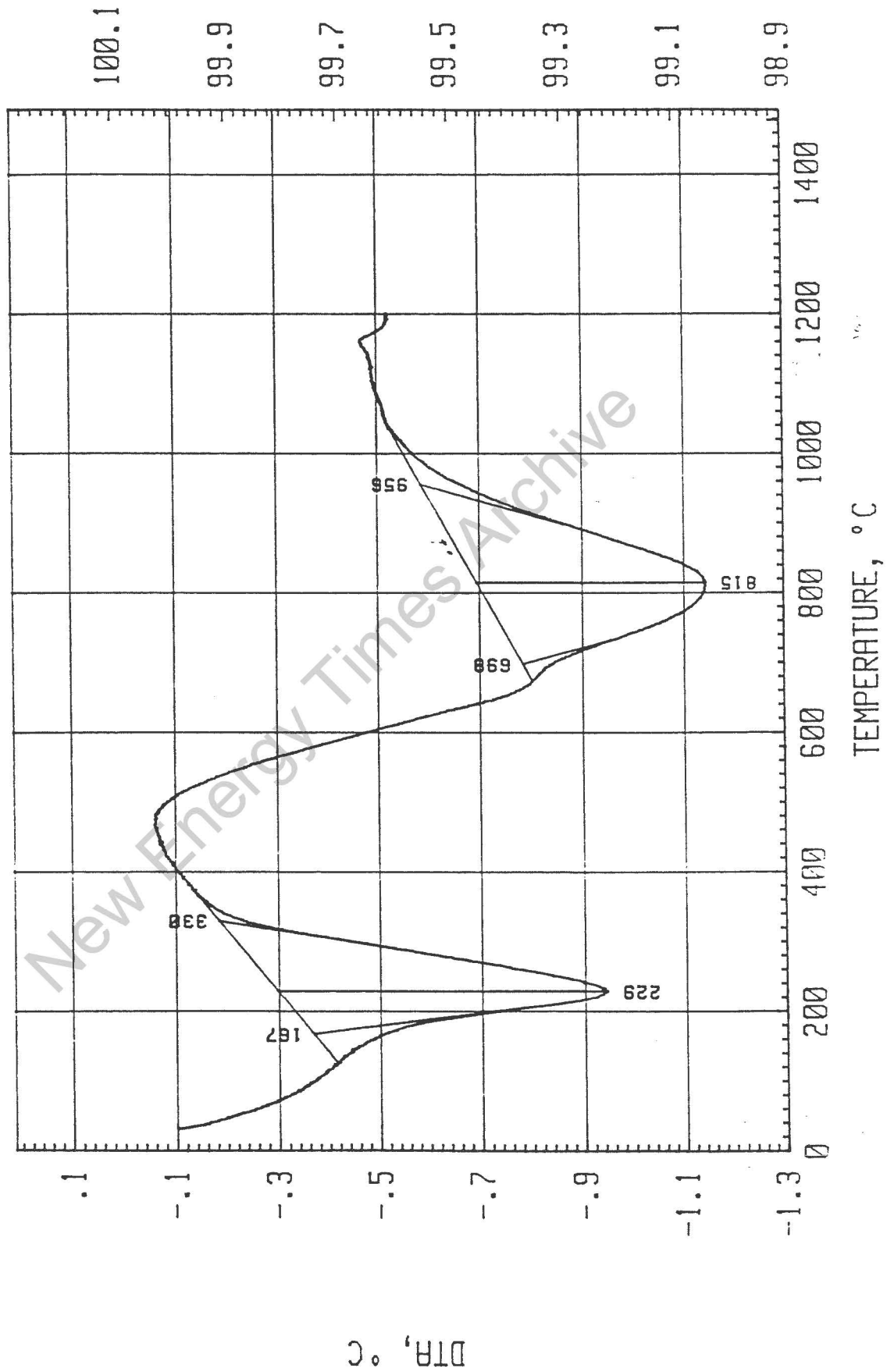
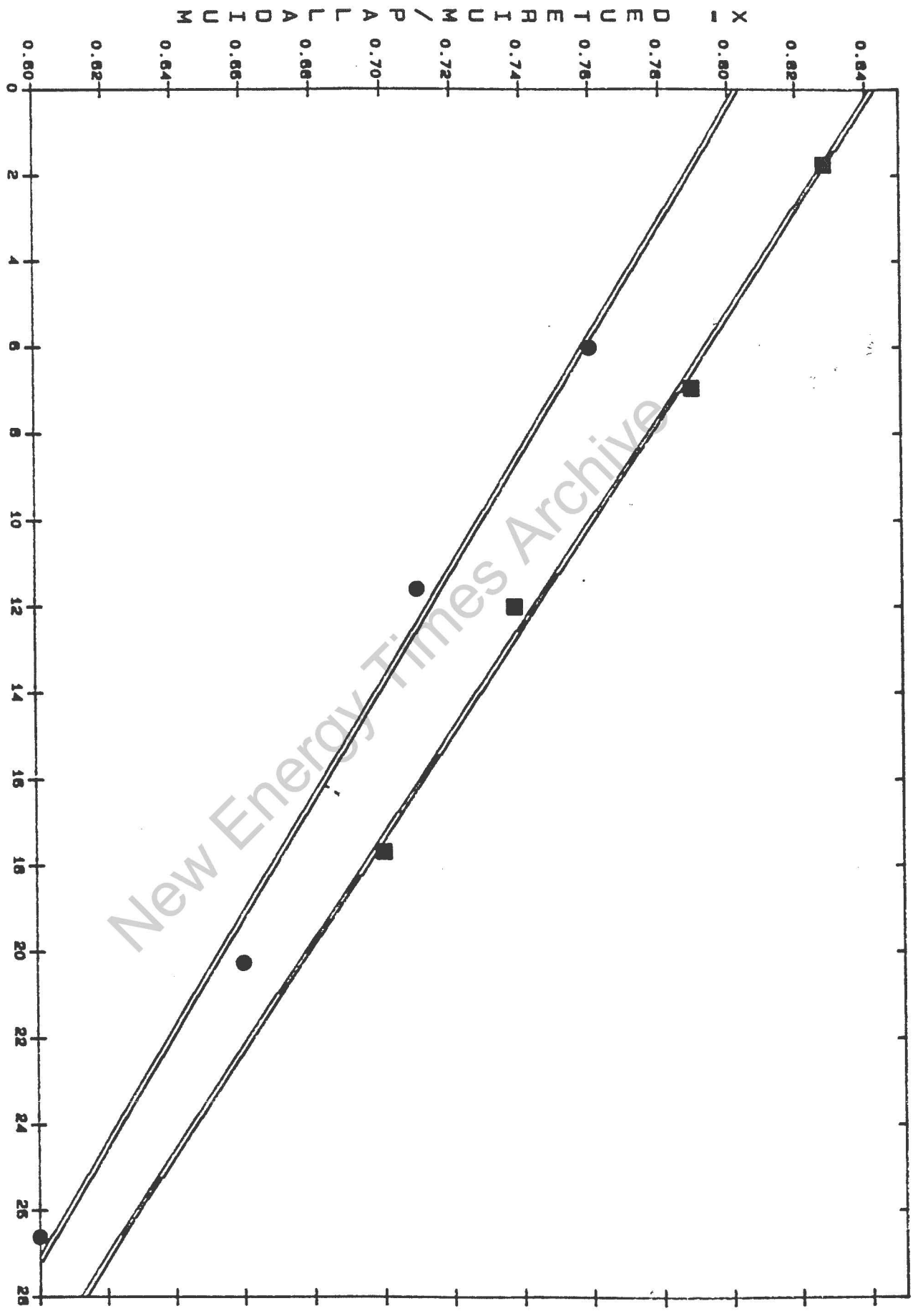


Fig. 4. THERMOGRAVIMETRIC ANALYSIS AND DIFFERENTIAL THERMAL ANALYSIS OF COLD CAST PALLADIUM ELECTRODE: NO PRETREATMENT, NO ELECTROCHARGING.

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SQUARE ROOT OF TIME (HOURS)



## VI. ACKNOWLEDGEMENTS

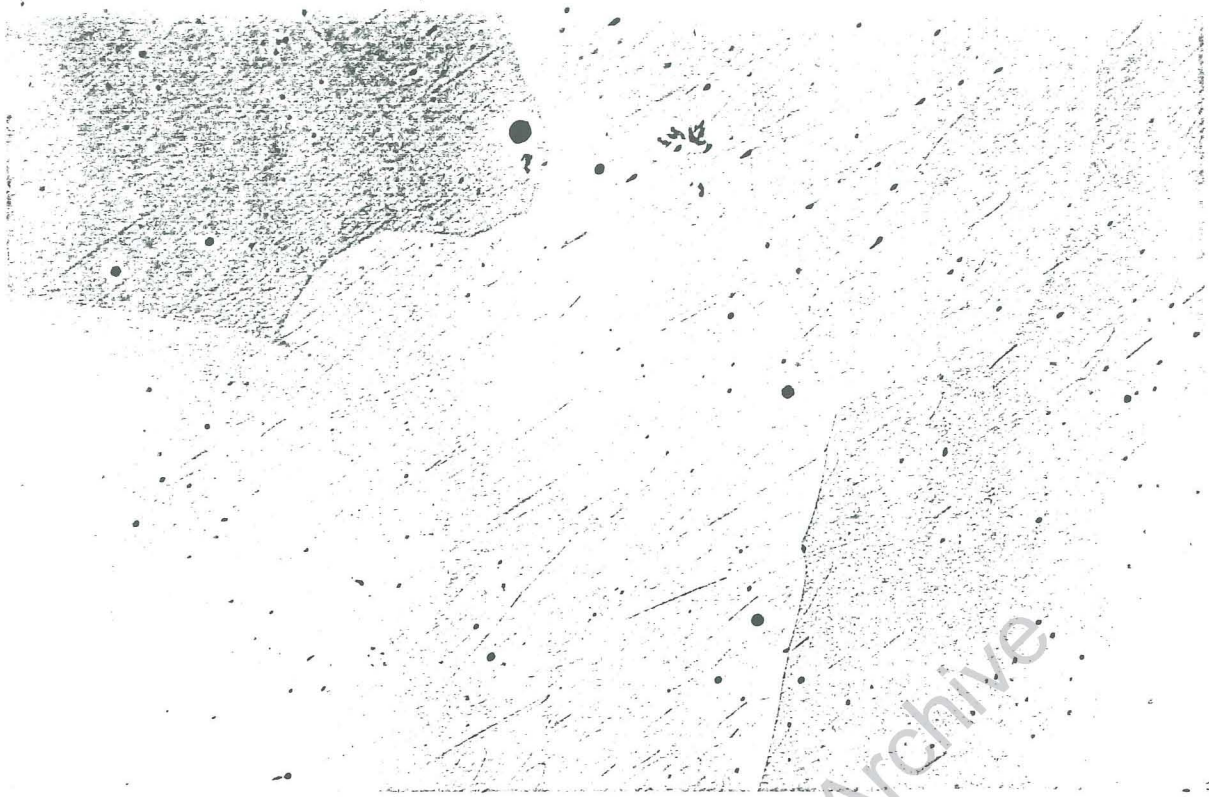
The authors are grateful for the efforts of the craftsmen of the Plant and Equipment Division of ORNL. The many details of power, plumbing, electronics, etc. that were addressed allowed this program to start smoothly and operate efficiently even when more established programs ostensibly had higher priority. We are also thankful for the many efforts of the technicians who are assigned to the Ceramic Preparations Group of the Metals and Ceramics Division: Claudia Walls, Clyde Hamby, Bill Chilcoat, Becky Freeny, and April Underwood. Although none were directly involved in experimental operations, they were helpful for a beaker here, a clamp there, etc. Never underestimate their ability to get a job done. We appreciate the valuable aid given to us by the ORNL Instruments and Controls Division in designing, fabricating, and providing measuring and counting equipment: Martin Bauer coordinated the efforts of a number of personell.

Without the backing of the management on every level we would not have been able to carry out this work. We feel that though many were not sure that "cold fusion" had any great amount of credence, the need for investigation was apparent to all.

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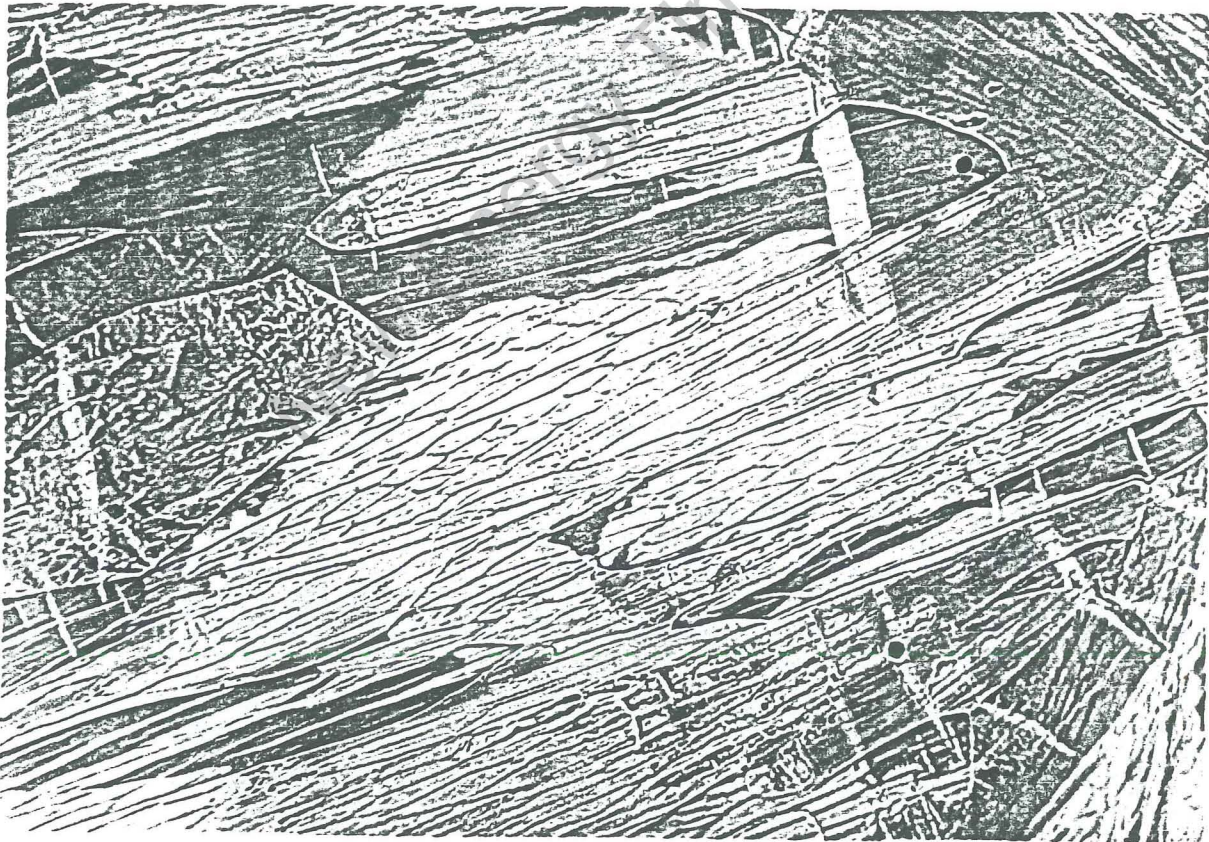
Fig. 11. KINETIC EVALUATION OF THE RELEASE OF DEUTERIUM FROM AN ELECTROCHARGED PALLADIUM CATHODES. Lower Curve: Cold cast palladium without annealing. Upper Curve: Hot cast palladium. Lines are linear least squares approximations for the data.

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200  $\mu\text{m}$

100X



200  $\mu\text{m}$

100X

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OPERATOR:  
DATE:  
RUN No.  
SAMPLE No.  
SAMPLE NAME:

WMS  
8 Aug 1989  
1 2  
1  
PD ELECT

\*\* NETZSCH STA ANALYSIS \*\*  
REFERENCE: AL203  
CRUCIBLE: AL203  
FILE NAME: B1:PD\_2  
LOT No. 1

ATMOSPHERE  
FLOW RATE, cc/min:  
HEAT RATE, °C/min:  
SAMPLE Wt., mg:  
REFERENCE Wt., mg:

HE  
75  
10  
273.7  
272.4

BUOYANCY



TG, % Sample Wt.

Fig. 10. METALLOGRAPHIC POLISHED SECTION OF PALLADIUM CATHODE: COLD CAST, NO ANNEALING, ELECTROCHARGED AND AIR EQUILBRATED FOR 22 DAYS. A. Observation shortly after electrocharging shows large homogeneous grains. B. Observation 22 days after electrocharging reveals considerable numbers of defects and periodic arrays of inhomogeneity.

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OPERATOR:		WMS		** NETZSCH STA ANALYSIS **		ATMOSPHERE		HE	
DATE:		8 Aug 1989		REFERENCE:		FLOW RATE, cc/min:		75	
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SAMPLE NAME:		PD ELECT		LOT No.		REFERENCE Wt., mg:		272.4	

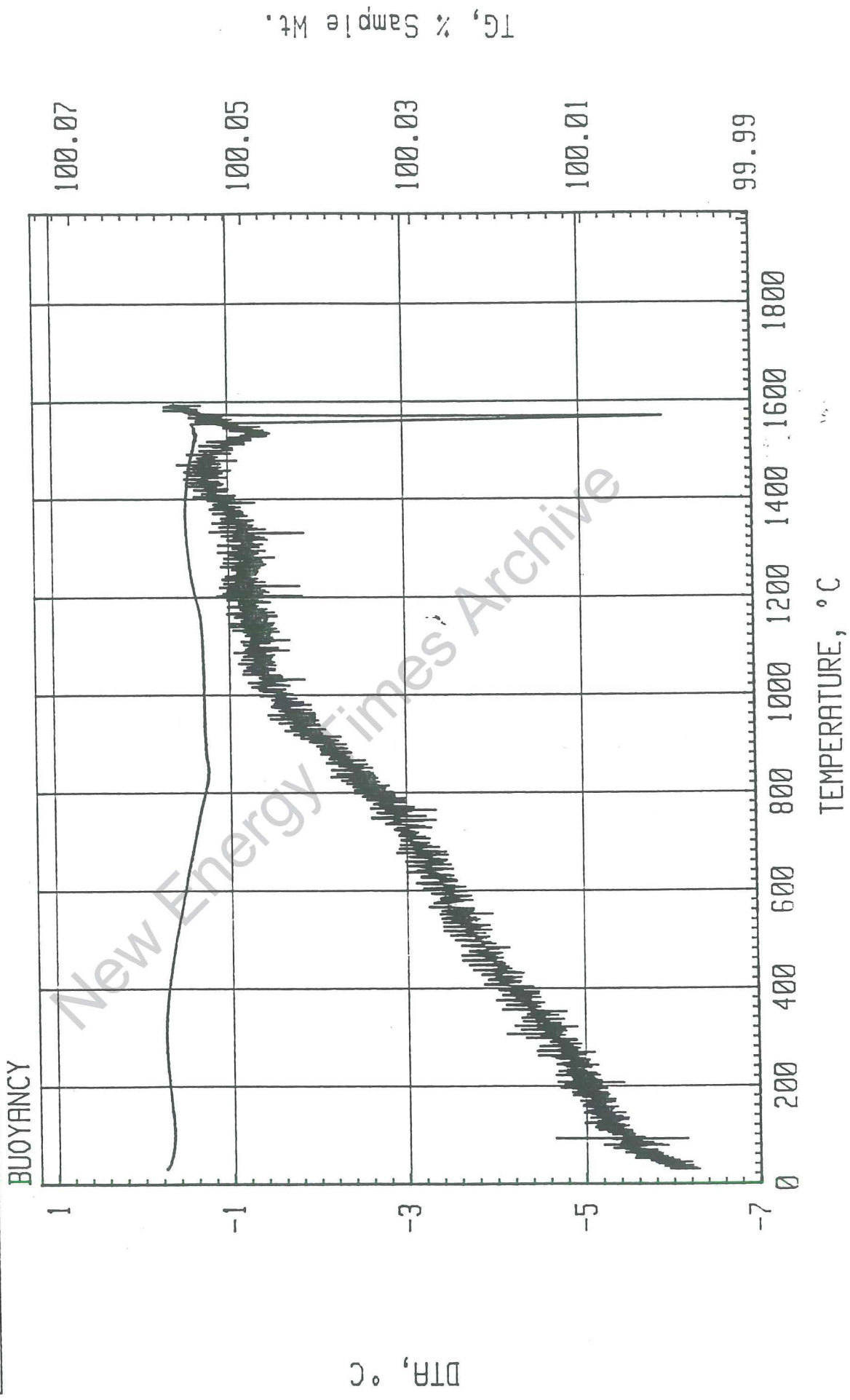


Fig. 8. EXPANDED DIFFERENTIAL THERMAL ANALYSIS OF THE MELTING OF COLD CAST PALLADIUM ELECTRODE AFTER ANNEALING: NO ELECTROCHARGING.

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OPERATOR:

DATE: 7 Aug 1989

RUN No.

SAMPLE No.

SAMPLE NAME:

HMS

1

1

PD ELECT

\*\* NETZSCH STA ANALYSIS \*\*

REFERENCE:

CRUCIBLE:

FILE NAME:

LOT No.

AL203

AL203

B1:PD STD

1

ATMOSPHERE

FLOW RATE, cc/min:

HEAT RATE, °C/min:

SAMPLE Wt., mg:

REFERENCE Wt., mg:

HE

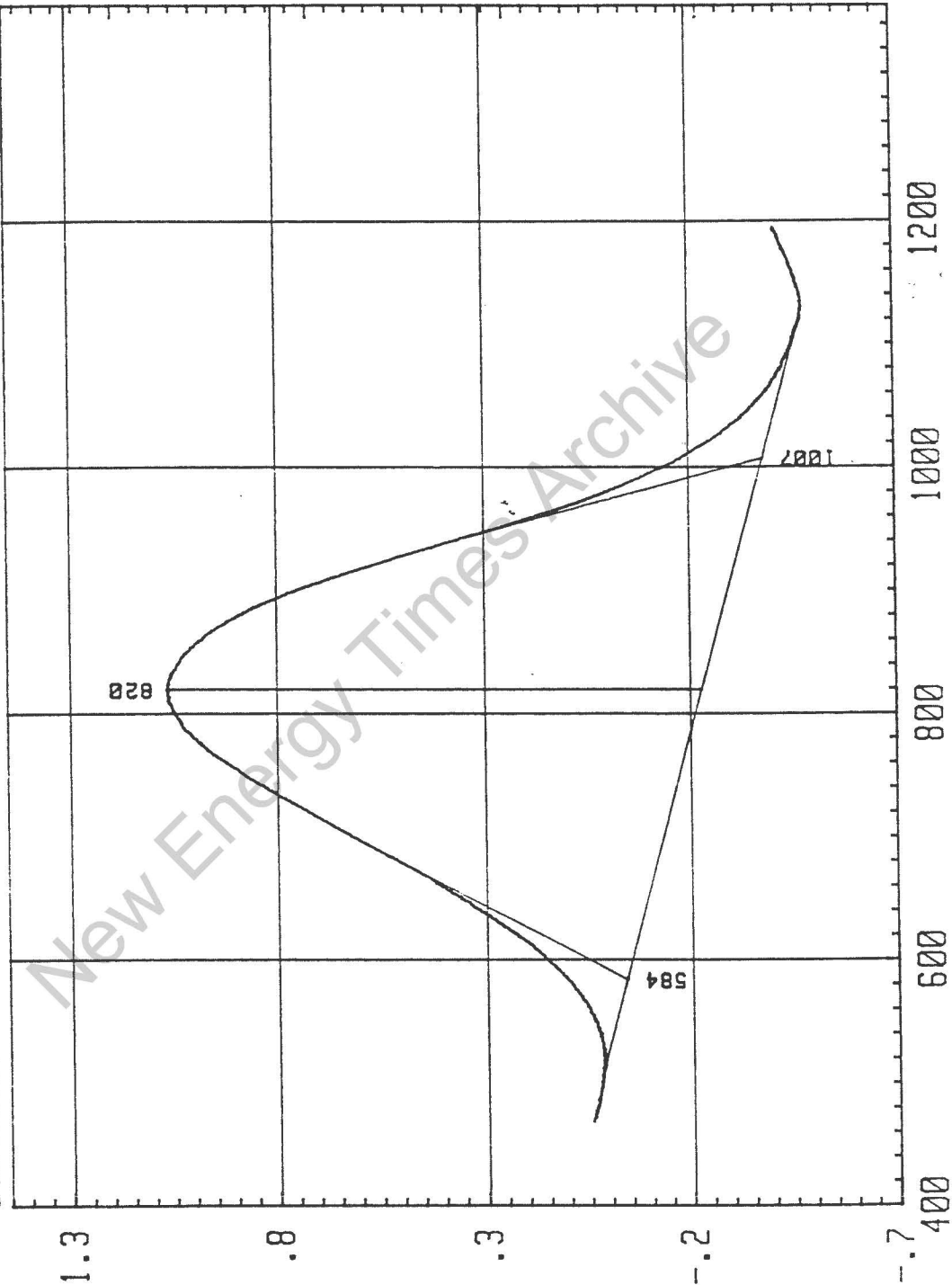
75

10

273.7

272.4

BUOYANCY



TG, % Sample Wt.

Fig. 7. THERMOGRAVIMETRIC ANALYSIS AND DIFFERENTIAL THERMAL ANALYSIS OF COLD CAST PALLADIUM ELECTRODE AFTER ANNEALING: NO ELECTROCHARGING.

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OPERATOR:	HMS	** NETZSCH STA ANALYSIS **		ATMOSPHERE	HE
DATE:	7 Aug 1989	REFERENCE:	AL203	FLOW RATE, cc/min:	75
RUN No.	1	CRUCIBLE:	AL203	HEAT RATE, °C/min:	10
SAMPLE No.	1	FILE NAME:	BI:PD STD	SAMPLE Wt., mg:	273.7
SAMPLE NAME:	PD ELECT	LOT No.	1	REFERENCE Wt., mg:	272.4

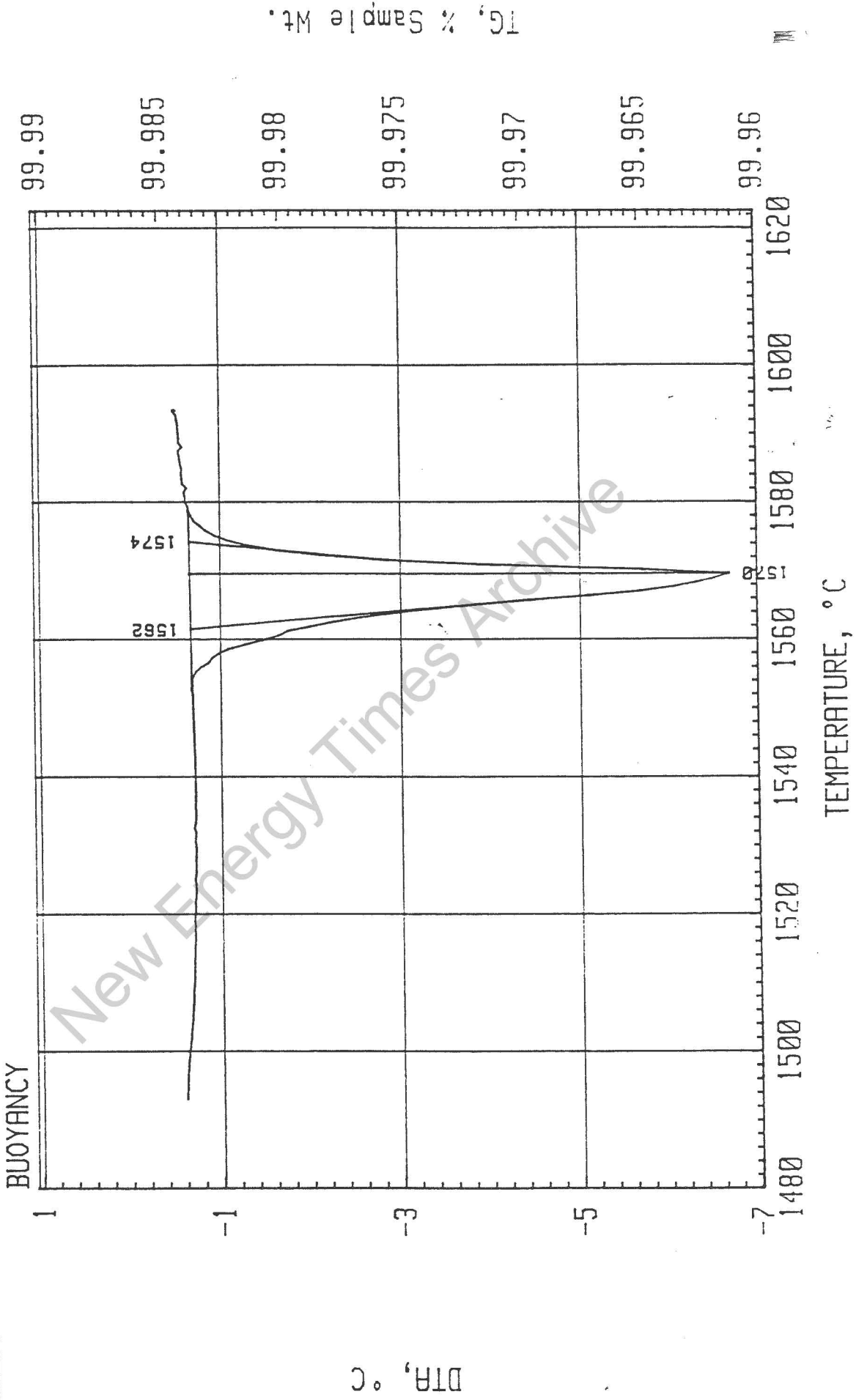


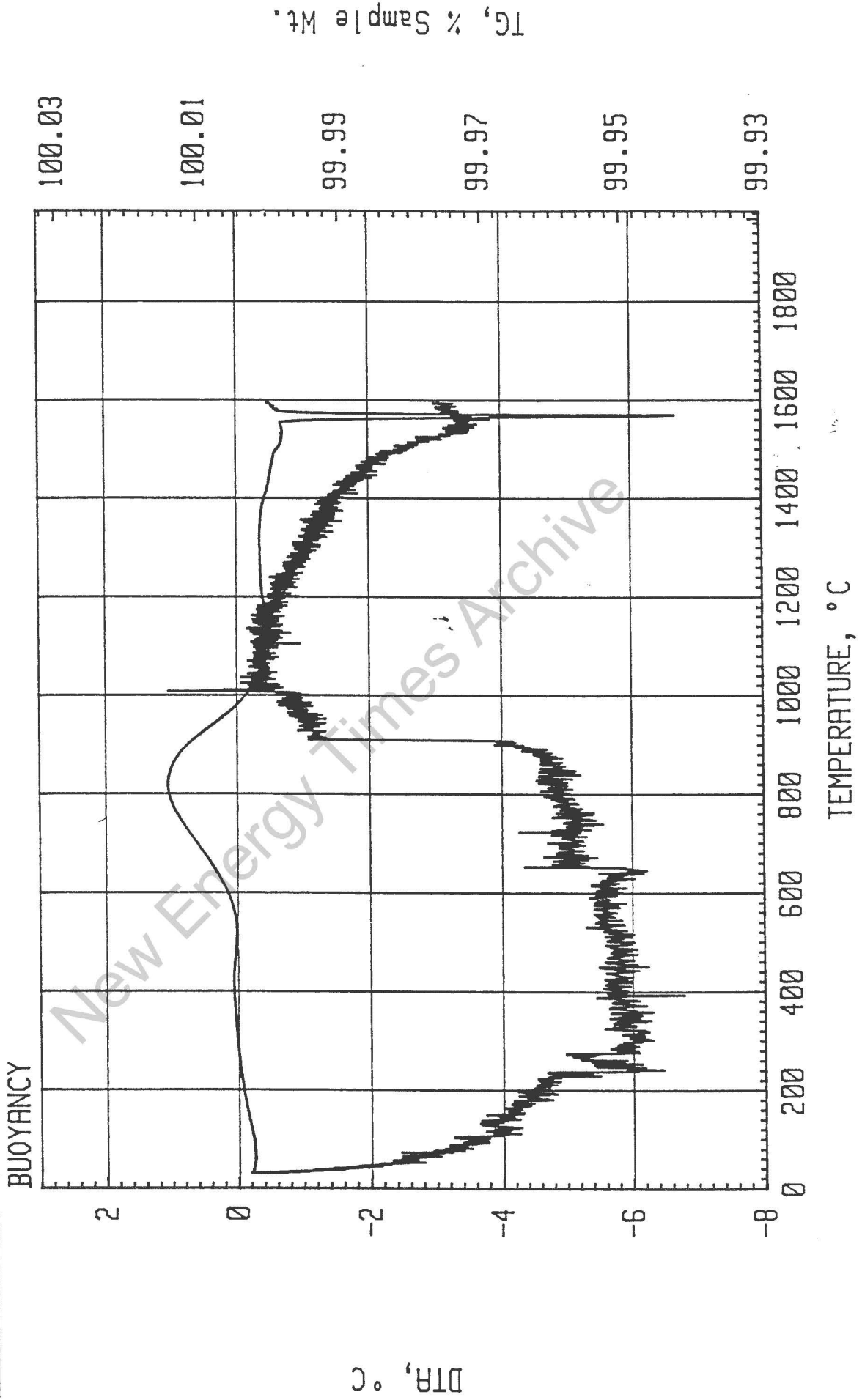
Fig. 6. EXPANDED DIFFERENTIAL THERMAL ANALYSIS OF THE ANNEALING OF COLD CAST PALLADIUM ELECTRODE: NO PRETREATMENT, NO ELECTROCHARGING.

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Fig. 5. EXPANDED DIFFERENTIAL THERMAL ANALYSIS OF THE MELTING OF COLD CAST PALLADIUM ELECTRODE: NO PRETREATMENT, NO ELECTROCHARGING.

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OPERATOR:	AMS	** NETZSCH STA ANALYSIS **		ATMOSPHERE	HE
DATE:	7 Aug 1989	REFERENCE:	AL203	FLOW RATE, cc/min:	75
RUN No.	1	CRUCIBLE:	AL203	HEAT RATE, °C/min:	10
SAMPLE No.	1	FILE NAME:	BI:PD STD	SAMPLE Wt., mg:	273.7
SAMPLE NAME:	PD ELECT	LOT No.	1	REFERENCE Wt., mg:	272.4



Tests for "Cold Fusion" in the Pd-D<sub>2</sub> and Ti-D<sub>2</sub> Systems  
at 40 to 380 MPa and -196 to 27°C

by

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## ABSTRACT

Experiments have been conducted on the Pd-D<sub>2</sub> and Ti-D<sub>2</sub> systems at 40 to 380 MPa and -196 to 27°C to investigate the possibility that "cold fusion" occurs in palladium and titanium deuterides generated by reaction with high-pressure D<sub>2</sub> gas. The experiments were performed using a 4.8 mm i.d. stainless steel pressure vessel that can be operated routinely at pressures as high as 400 MPa.

In experiments completed to date, reactions between high-purity Pd or Ti and D<sub>2</sub> were monitored with: (1) an array of three BF<sub>3</sub> neutron detectors; (2) an internal, type-K sample thermocouple; and (3) an internal, type-K reference thermocouple located approximately 10 cm above the sample thermocouple. Using a <sup>252</sup>Cf source, the efficiency of the BF<sub>3</sub> detector array was determined to be approximately 6%. During experimentation, the three neutron detectors were immersed in a water bath thermostated at 27°C. The neutron count rate, D<sub>2</sub> pressure, sample and reference thermocouple readings, and bath temperature were recorded continuously at time intervals ranging from 6 seconds to 10 minutes.

Experimental results obtained so far range from negative to potentially significant. No sustained heat production has been observed in any experiment. Thermal pulses that persist briefly after pressurizing Pd with D<sub>2</sub> gas are attributable to small amounts of chemical heat released when Pd and D<sub>2</sub> react to form palladium deuteride. No sustained neutron flux above background was observed in any Pd-D<sub>2</sub> experiment.

On the other hand, in a Ti-D<sub>2</sub> experiment just completed, potentially significant results were obtained. During this experiment, there was a period

of 5 consecutive hours when count rates rose to approximately 60 counts/hour above the average background rate. This detector count rate corresponds nominally to 1000 neutrons/hour emitted from the Ti-D<sub>2</sub> sample. However, due to several deficiencies in our neutron detection methods and equipment, we cannot demonstrate conclusively that our experimental data are valid. Consequently, we are upgrading our neutron detection equipment in preparation for a second, improved Ti-D<sub>2</sub> experiment.

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## INTRODUCTION

In late March of this year, Stanley Pons of the University of Utah and Martin Fleischmann of the University of Southampton announced that they had induced "cold fusion" of deuterium atoms by electrochemically loading deuterium into palladium cathodes in an LiOD-D<sub>2</sub>O electrolyte at room temperature. The evidence for this claim was an apparent excess heat output above the energy required to sustain electrolysis (the "Fleischmann and Pons effect," Fleischmann and Pons, 1989). These authors also claimed to have detected fusion products - 2.4 MeV neutrons and tritium - in their experiments, although at levels many orders of magnitude below that required to explain the excess heat. At about the same time, Steven Jones of Brigham Young University reported extremely low levels of 2.4 MeV neutrons emitted from electrochemical cells using a complex electrolyte mixture in D<sub>2</sub>O, including LiOD, with either palladium or titanium cathodes (Jones et al., 1989). These announcements generated great excitement in the scientific community, with many research groups attempting to reproduce and explain the phenomena.

As subsequent developments concerning "cold fusion" were reported in press conferences, news accounts and interlaboratory communications, it became apparent that many researchers believed that cold fusion was induced by loading high concentrations of deuterium into a suitable metal - palladium, in particular. Accordingly, we decided to attempt to achieve such loading by exposing palladium to high-pressure D<sub>2</sub> gas. The experimental setup we envisioned was a pressure vessel immersed in a thermostated water bath, with thermal effects and fusion phenomena monitored using thermocouples and BF<sub>3</sub>

neutron detectors. A schematic representation of this experimental design is shown in Figure 1.

Shortly after we initiated our Pd-D<sub>2</sub> experiments, we learned of the work of De Ninno et al. (1989) at the Institute for Alternative Energy Resources at Frascati in Italy. The Frascati experiments involved exposing titanium turnings to -4 MPa of D<sub>2</sub> gas in a pressure vessel placed near a BF<sub>3</sub> neutron detector. When cooled down to -196°C with liquid nitrogen and allowed to warm back up to room temperature, the pressurized titanium was reported to emit bursts of neutrons at levels up to 10<sup>5</sup> times that observed by Jones et al. (1989). This release of neutrons apparently could be reproduced many times with a single sample. The Frascati group also reported the results of an experiment in which a Ti-D<sub>2</sub> sample was depressurized while warming up from -196°C. In this experiment, vacuum pumping and warming of the sample were initiated simultaneously and, 3 hours thereafter, the titanium apparently began emitting neutrons at a rapid rate (reportedly up to 5000 neutrons/second). This neutron-emission event continued for approximately 13 hours. The foregoing experimental observations of De Ninno et al. (1989) are referred to collectively as the "Frascati effect." To investigate the Frascati claims, we also conducted a high-pressure Ti-D<sub>2</sub> experiment.

#### EXPERIMENTAL EQUIPMENT

The stainless steel pressure vessel shown in Figure 1 has a 4.8 mm internal diameter, a 7.9 mm external diameter, and is 30.5 cm long. Type-K thermocouples are sealed in place with silver solder, and the annular space around the upper (reference) thermocouple allows introduction or removal of

gas. The pressure vessel is connected to a hydrogen-service, high-pressure gas intensifier that is capable of generating 400 MPa of  $H_2$  or  $D_2$  gas pressure. The neutron detector array consists of three 5.1 cm o.d. x 40.6 cm  $BF_3$  tubes (Reuter-Stokes #44A) arranged radially around the vessel and operated in parallel at 1900 volts. The detector system has an efficiency of ~6%, as determined using a  $^{252}Cf$  source (Figure 2). The background count rate of the  $BF_3$  detectors is approximately 9.5 counts/minute, which yields a sensitivity at the  $3\sigma$  level of about 1200 neutrons/hour from the experimental sample. The vessel and detectors are immersed in a thermostated water bath to maintain constant temperature and to thermalize fast neutrons produced by cold fusion.

#### DATA ON THE Pd- $D_2$ SYSTEM

A series of experiments were conducted to elucidate reactions between Pd and  $D_2$  at high pressures and low temperatures. In our initial Pd- $D_2$  experiment, 0.7 grams of 0.1-mm thick palladium ribbon (Table 1) was wrapped around the tip of the lower (sample) thermocouple and sheathed with platinum foil. Pressurization of this sample to 380 MPa with 99.87%  $D_2$  gas produced a rapid temperature rise (from 27 to 60°C) that was much larger than a simultaneous temperature rise (from 27 to 32°C) recorded by the reference thermocouple. This thermal anomaly decayed to the bath temperature after 12 minutes. During and for 5 days after this pressurization, the neutron flux was monitored at sampling times ranging from 6 seconds to 10 minutes. No sustained neutron flux above background was observed. Subsequent pressurizations of  $D_2$  gas alone and 0.7 grams of palladium with  $H_2$  gas produced thermal effects similar to the initial run, indicating that the thermal anomaly observed during the

Pd-D<sub>2</sub> experiment can be attributed to (1) PV work accompanying gas pressurization, (2) heat released during the formation of palladium deuteride, and (3) the somewhat different geometries of the sample and reference thermocouples.

The pressure vessel was then packed with 7.0 grams of palladium in the form of (1) ~1 gram of 0.1-mm thick ribbon wrapped around the sample thermocouple, and (2) ~6 grams of 3 mm x 3 mm cut pieces of 0.1-mm thick ribbon filling the space between the sample and reference thermocouples (Figure 1). Upon initial pressurization to 380 MPa with D<sub>2</sub> gas, thermal effects similar to those described above were observed. During the next 3 days, the temperature of the vessel was (1) reduced to -78°C using dry ice, and subsequently (2) allowed to warm slowly to room temperature. This was done because several investigators (e.g., Perminov et al., 1952) have reported greatly enhanced solubility of hydrogen in palladium at cryogenic temperatures. Finally, the sample was depressurized to a 20 micron vacuum at room temperature for one day and repressurized to 350 MPa for two days. As in the first Pd-D<sub>2</sub> experiment, no sustained neutron flux above background was observed.

#### DATA ON THE Ti-D<sub>2</sub> SYSTEM

A single Ti-D<sub>2</sub> experiment has been conducted with the same high-pressure equipment that was used to perform the Pd-D<sub>2</sub> experiments described above. Furthermore, experimental methods employed in the Ti-D<sub>2</sub> experiment were similar to those used in the Pd-D<sub>2</sub> experiments. The principal goal of the Ti-D<sub>2</sub> experiment was to test the claim of De Ninno et al. (1989) that "disequilibrium conditions" would induce cold fusion in Ti-D<sub>2</sub> samples.

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## FIGURE CAPTIONS

Figure 1

A schematic illustration of the pressure vessel and array of  $\text{BF}_3$  detectors used to perform cold fusion experiments.

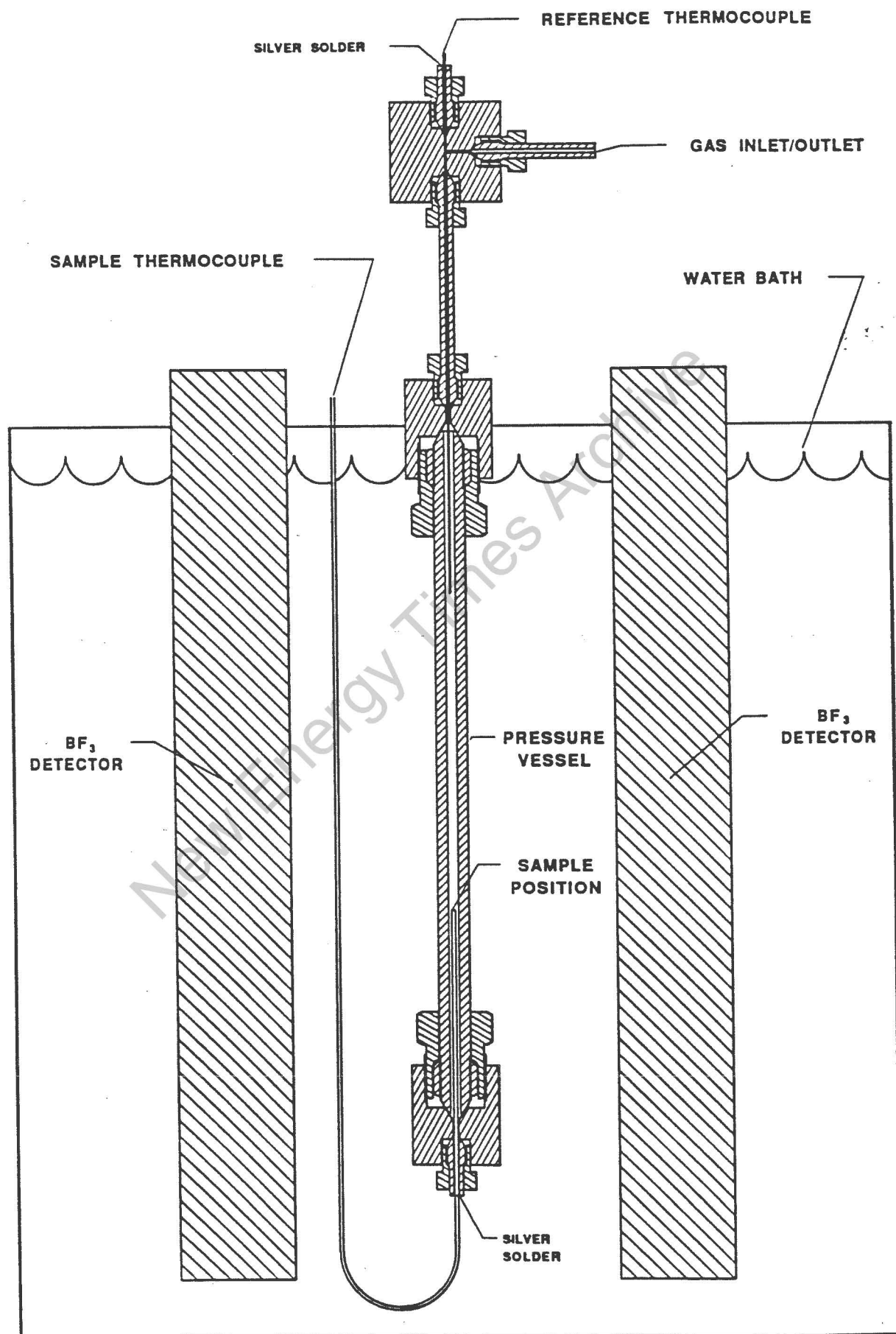
Figure 2

Pulse height spectrum from a  $^{252}\text{Cf}$  source (500 neutrons/second). Noise pulses below channel 20 were suppressed. For neutron counting, the integral bias was set at channel 58. The error bars illustrate  $1\sigma$  values for individual data points.

Figure 3

Variations in pressure (MPa), temperature ( $^{\circ}\text{C}$ ) and detector count rate (counts/hour) during our Ti- $\text{D}_2$  experiment. Lower diagram: pressure (dashed lines) and temperature (solid lines) plotted against time in hours. Upper diagram: detector count rate plotted against time in hours. The error bars illustrate  $1\sigma$  values for individual data points.

Figure 1



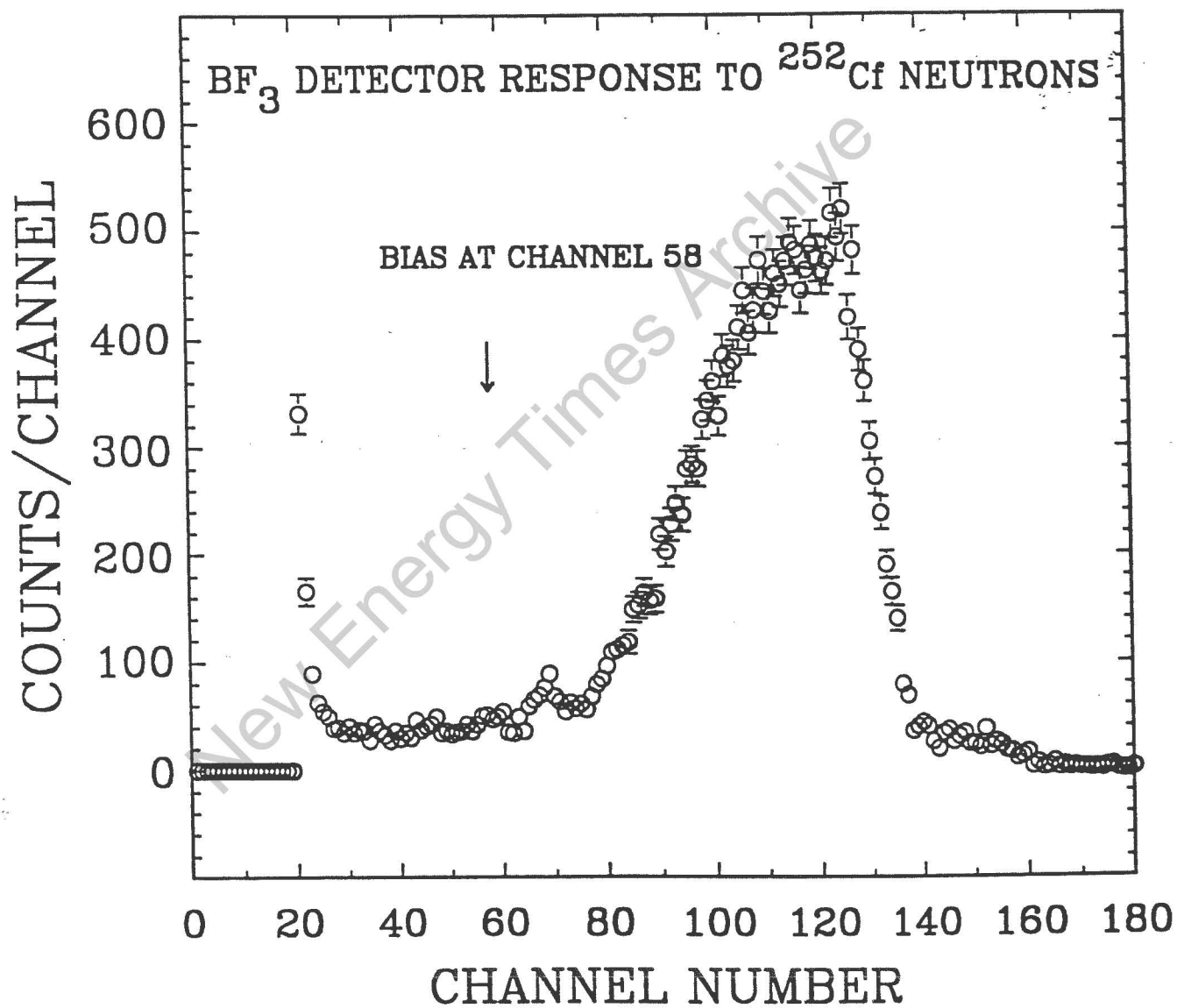


Figure 3

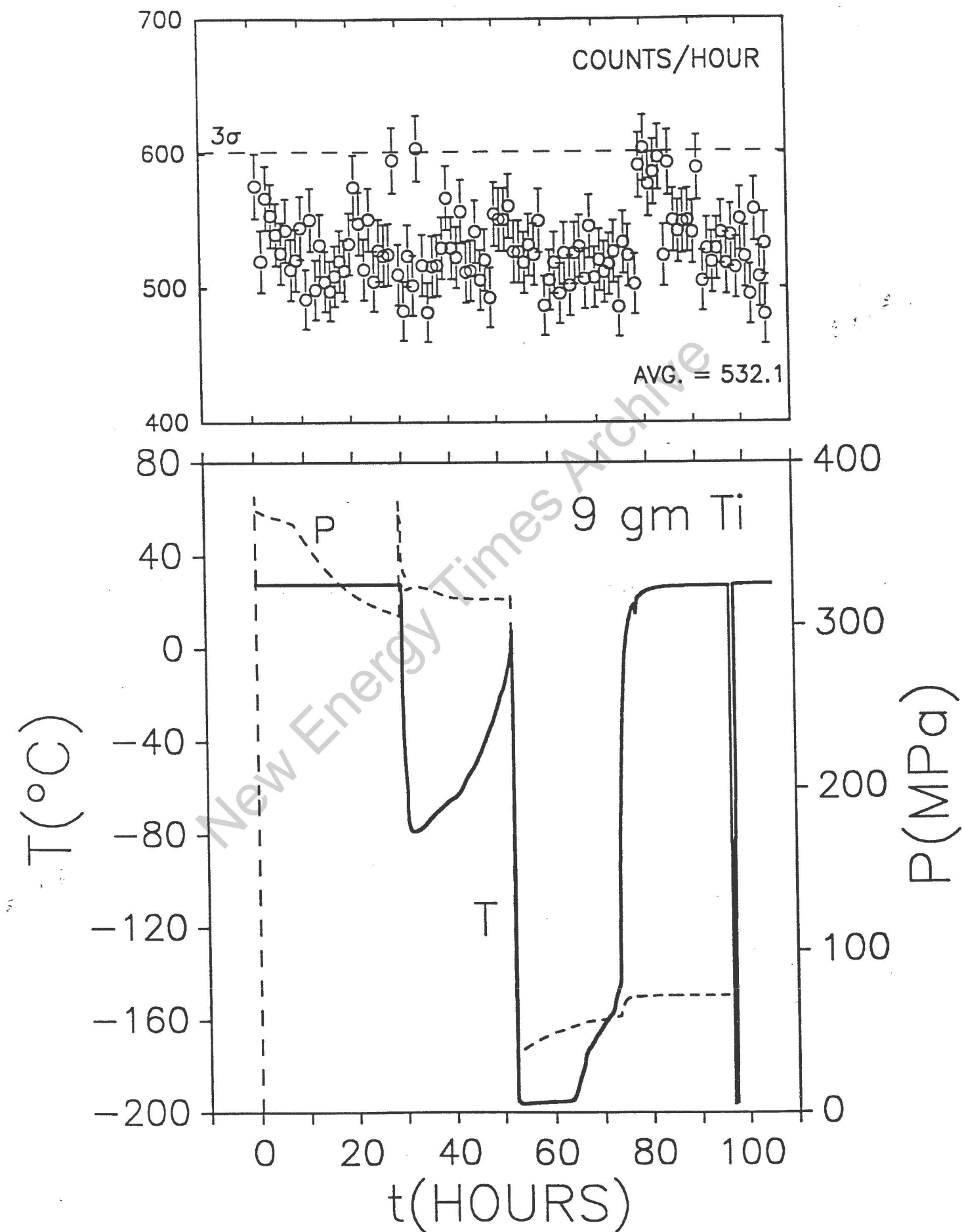


Table 1. Compositions of titanium and palladium used in this study.<sup>a</sup>

Analysis of 99.97% Palladium Ribbon <sup>b</sup>							
Ag 6	As 0.7	Au 1	B 12	Ba <.2	Be .20	Bi <0.1	
Br <.04	Ca 73	Cd <.20	Co .02	Cs <.03	Cu 9	Fe 19	
Ga <.03	Ge <.02	Hf <.05	Hg <.08	In <.01	Ir 3	K .50	
Mg .70	Mo 2	Na .07	Nb .20	Ni 3	Os <.05	P .30	
Pb .7	Pt 52	Rb <.01	Re <.02	Rh 29	Ru <.06	S 3	
Sb <.05	Sc .02	Se <.01	Si 54	Sn <.30	Sr .03	Ta <5	
Te 5	Th .06	Ti 2	Tl <.02	U .20	W .60	Y <.02	
Zn 2	Zr 4	Cl .40	F <.01	I <.03	Ce <.10	Dy <.04	
Er <.04	Eu <.02	Gd <.05	Ho <.01	La <.04	Lu <.01	Nd <.05	
Pr <.01	Sm <.04	Tb <.01	Tm <.01	Yb <.04			

Analysis of 99.91% Titanium Wire <sup>c</sup>							
C 35.0	N 15.0	O 500.0	Mg <5.0	Si <5.0	Al 20.0	S 10.0	
Ca 5.0	V 5.0	Cr 40.0	Mn 30.0	Fe 150.0	Ni 40.0	Cu 30.0	
Sn 10.0	Pb 20.0	Bi <10.0					

<sup>a</sup>Concentrations of all trace elements reported in ppm.

<sup>b</sup>Analyzed by DC-arc spectrograph at the Y-12 Plant, Oak Ridge, TN. Masked elements include Al, Cr, Mn, and V. Prior to use in experimentation, the ribbon was: cleaned in concentrated aqua regia; heated to 600°C under high vacuum ( $10^{-5}$  torr); exposed to 0.05 MPa D<sub>2</sub> gas at 600°C for 15 minutes; held at 600°C under high vacuum ( $10^{-5}$  torr) for one hour; cooled back to room temperature; and finally, stored under 99.999% argon. Additionally, to preclude surface contamination prior to experimentation, the ribbon was loaded into the pressure vessel under 99.999% argon.

<sup>c</sup>Lot analysis reported by vendor (#22/58107, Materials Research Corp., Orangeburg, NY). Prior to use in experimentation, the wire was chemically polished in an acid bath (nitric + lactic + hydrofluoric acid in a 5:5:4 ratio) to remove all surficial titanium oxide.

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# PRELIMINARY INVESTIGATION OF POSSIBLE LOW-TEMPERATURE FUSION

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\*Operated by Martin Marietta Energy Systems, Inc., under contract DE-AC05-84OR21400 with the U.S. Department of Energy.

# PRELIMINARY INVESTIGATION OF POSSIBLE LOW-TEMPERATURE FUSION

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## ABSTRACT

Preliminary tests have been made with electrolytic cells utilizing 0.2 N LiOD in D<sub>2</sub>O as the electrolyte and a palladium cathode surrounded by a wire-wound platinum anode operating at cathode current densities of 100 to 400 mA/cm<sup>2</sup>. The cathodes were swaged to diameters of 2.8 or 5.5 mm with 8.5 cm of active length. The electrolyte temperature was controlled, heat was removed by flowing water in a cooling jacket, and the cell was insulated. Cooling water and electrolyte temperatures were measured by thermocouples, and neutron and gamma-ray spectra were recorded. The electrolyte level was periodically monitored and replenished with D<sub>2</sub>O. Tests up to 2 weeks in duration were made with no sustained release of energy in excess of the electrical power input, although there was one period of 12 h when an unaccountable heat excess was observed. In another test, an anomalous neutron flux was measured during the first few hours that was 3.5 standard deviations above the background.

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\*Operated by Martin Marietta Energy Systems, Inc., under Contract DE-AC05-84OR21400 with the U.S. Department of Energy.

**KEYWORDS;** Cold fusion, electrolysis, palladium cathode

## 1. INTRODUCTION

There have been recent experimental indications from several research groups that excess energy has been observed during the electrolysis of heavy water in a LiOD electrolyte solution with a palladium cathode.<sup>(1-3)</sup> In some cases, neutron flux anomalies have been observed and an increase in tritium content of the electrolyte solution has also been measured.<sup>(3-5)</sup>

A group of researchers at Oak Ridge National Laboratory, which includes chemists, physicists and chemical engineers, has initiated an investigation with the goal of experimentally studying possible thermal or nuclear reactions occurring during electrolysis under the above conditions. The experimental system utilized positive heat removal by circulating cooling water, a design concept that is amenable to system scaleup and continuous operation.

## 2. MATERIALS AND METHODS

### 2.1. Electrochemical Cell

The electrolysis cell was fabricated from Pyrex glass with a nominal ID of 4 cm and an active internal height of 12.5 cm (Fig. 1). There was also an internal head space of  $\sim 4$  cm in height which was continuously purged by  $N_2$  gas. A Teflon cap was used to partially seal the system and support electrodes, an electrolyte thermocouple encased in glass, a glass-encased heating source for internal calibration, and a polyethylene tube for introduction of make-up water and removal of electrolyte samples. The cell was encased in a cooling jacket with forced water flow and thermocouples in the inlet and outlet streams. Two inches of fiber glass insulation was used on the entire exterior of the cell except for a 1- x 3-cm opening in the insulation that was used to observe the electrolyte level.

Cathode rods of 99.95% palladium were prepared by casting under argon and then swaging to the final dimensions of 0.28- or 0.55-cm diam x 8 to 9 cm. Electrical contact was via a 0.050-in. Pt wire, electron-beam welded to the Pd cathode. The anodes were 0.02-in.-diam Pt wire

that was wound around a glass skeletal mandrel which surrounded the cathode with an annular electrode separation of 0.4 to 0.5 cm. The cell was operated at a cathode current density of 100 to 400 mA/cm<sup>2</sup>.

## 2.2. System Design

Heat removal was determined by the temperature increase of the forced flow of cooling water through the jacketed cell. A circulating coolant bath with temperature controlled to within  $\pm 0.1^{\circ}\text{C}$  was used with an external positive displacement pump that controlled the coolant flow rate with an accuracy of 2 to 4% (Fig. 2). Rotameters were employed to monitor the coolant and N<sub>2</sub> purge gas flow rates. Heavy water replenishment and electrolyte sampling were performed intermittently by means of a manually operated syringe.

The potentiostat/galvanostat was operated in constant current mode with an accuracy of  $\pm 1$  mA. Electrolyte and coolant water inlet and outlet temperatures were measured by calibrated thermocouples with an accuracy of  $\pm 0.1^{\circ}\text{C}$ , and the overall electrode voltage was measured to an accuracy of  $\pm 0.01$  V. These system parameters were recorded on a strip chart recorder every 6 s.

## 2.3. Radiation Detection

The experimental system was located within a 2-ft-thick concrete enclosure in order to reduce background radiation. Neutrons were measured by a NE-213 scintillator placed immediately adjacent to the insulated cell (Fig. 3). This detector employed pulse-shape discrimination to differentiate between neutrons and gamma rays and the threshold energy for fast neutron detection was set at 1.2 MeV. The overall neutron detection efficiency of  $1.46 \times 10^{-3}$  was determined by a <sup>252</sup>Cf source. Gamma-ray contribution to the neutron peak was 4.1% of the gamma peak or  $\sim 20\%$  of the neutron peak. The reported neutron flux was corrected for this contribution. This neutron detector had a typical sensitivity at 3 standard deviations of  $3 \times 10^{-24}$  fusions/deuterium-pair-s.

A separate NaI gamma-ray spectrometer with a polyethylene converter to generate capture gamma rays was also used for detection of neutrons. This system was lead-shielded and contained in an airtight enclosure to further reduce the background gamma-ray spectrum. A multichannel analyzer with a window set over the expected capture gamma-ray region was used; the system had an overall detection efficiency of  $5.75 \times 10^{-5}$  as determined by a  $^{252}\text{Cf}$  source. However, this system had somewhat less sensitivity due to the higher gamma-ray background. The tritium concentration in the electrolyte was measured periodically by an external scintillation system.

#### 2.4. Chemicals

The heavy water was 99.5%  $\text{D}_2\text{O}$  with an initial tritium content of  $1 \times 10^3$  to  $1.3 \times 10^4$  Bq/mL. The electrolyte was prepared by dissolving reagent-grade, natural lithium in the  $\text{D}_2\text{O}$  at a nominal concentration of 0.2 N.

#### 2.5. Operating Procedures

The tests were initiated by loading the prepared electrolytic cell with  $\sim 125$  mL of the electrolyte, starting the purge gas at  $\sim 1$  mL/s and turning on the electrical current. The electrolyte temperature was controlled by the cooling water temperature and flow rate and usually maintained in the range of 28 to 32°C with some controlled excursions to 70°C.

The various temperatures and the cell voltage were recorded every 6 s on an analog recorder and at 1-h intervals as digital displays. Heavy water ( $\text{D}_2\text{O}$ ) was replenished every 8 h in order to maintain a constant electrolyte concentration, and electrolyte samples were taken periodically (every 2 to 3 d). The gamma-ray and neutron spectra were measured continuously and recorded on magnetic media every 4 h.

## 2.6. Energy Balance

An energy balance was determined for each test based on the following assumptions:

(1) the electric current is 100% efficient for the electrolysis of  $D_2O$ ; (2) the system operates at quasi-steady state with a constant inventory of deuterium in the cathode; (3) all of the  $D_2$  and  $O_2$  exit the electrolysis cell without recombination; (4) the exiting gases, including the purge gas, were saturated with  $D_2O$  that was at equilibrium with heavy water at the temperature of the electrolyte; and (5) there was no heat loss to the ambient environment.

The second assumption will obviously not be correct during the initial phase of the experiment when a major portion of the formed  $D_2$  is being adsorbed by the cathode. Since the amount of make-up heavy water required to maintain a constant inventory in the electrolysis cell was approximately equal to that lost by electrolysis and evaporation, there was apparently very little recombination of the product gases. There was obviously some heat loss to the ambient although the system was well insulated, but this would result in a conservative estimate of the recovered heat.

The resulting energy balance can be represented by:

ENERGY IN:

Voltage (V) x Current (A)

ENERGY OUT:

1. ELECTROLYSIS (Typically >30%)

$D_2O \rightarrow D_2 + 1/2 O_2$  (-Heat of formation)

2. FORCED COOLING (Typically >60%)

(Temperature increase) x (Flow rate)

3. LATENT HEAT (Typically <2%)

(Heat of  $D_2O$  vaporization)

Chemical and physical properties for  $D_2O$  were obtained from reference handbooks.<sup>47</sup>

### 3. RESULTS AND DISCUSSION

The results of these tests are very preliminary and certainly do not confirm "cold fusion" as described by Pons and Fleischmann. However, during one short time period there was an apparent neutron increase, and during a period of a few hours there was apparently a significant amount of excess heat generated.

#### 3.1. Neutron Detection

In one test (CF-1) in which a palladium cathode 0.28-cm diam x 8.5 cm was used at a current density of 100 mA/cm<sup>2</sup>, the neutron background averaged 13.1 neutrons/4 h with a standard deviation of 5.3 neutrons/4 h. During the first few hours of this test there was a 4-h observation period, in which the neutron detector registered 32 counts/4 h (Fig. 4) exceeding the background count rate by 3.5 standard deviations. A frequency histogram of the neutron count data (Fig. 5), shows that the variability of our neutron observations for this run fits a binomial distribution with the possible exception of the anomalously high datum of 32 counts/4 h.

#### 3.2. Excess Heat

There was no sustained excess energy measured during either of the two tests. However, there were several short periods of time with apparent excess energy and one period of 12 h in one test when the energy recovered was in excess of the instantaneous input energy by as much as 50% (Fig. 6). That energy excess occurred during the second test (CF-2) with a 0.55-cm diam x 8-cm palladium cathode operating with a current density of 100 mA/cm<sup>2</sup>.

### 3.3. Additional Observations

The periodic addition of  $D_2O$  to maintain the heavy water inventory resulted in fluctuations of LiOD concentration and, thus, unsteady state operation since cell voltage changed with electrolyte concentration. A series of tests were also made at different electrolyte temperatures over the range of 30 to 70°C without apparent effect. Small tritium increases were measured in the electrolyte, but these did not exceed the expected increase due to the evaporation and electrode processes.

## 4. CONCLUSIONS

Preliminary tests of the electrolysis of  $D_2O$  utilizing LiOD electrolytes and palladium electrodes have not confirmed the "cold fusion" phenomena. However, there has been one apparently anomalous neutron flux measurement and periods of up to 12 h of apparent excess energy. None of these results have been reproduced nor can they be explained by conventional nuclear or chemical theory.

## 5. ACKNOWLEDGMENTS

We appreciate the assistance of H. E. Harmon in the preparation of the cathode materials; of J. W. Wade and colleagues for tritium analyses; and of J. E. Bigelow, P. W. Fisher, M. L. Bauer, and M. M. Chiles for their valuable consultations.

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## FIGURE CAPTIONS

Fig. 1. Electrolysis cell used in cold fusion study.

Fig. 2. Schematic of experimental system.

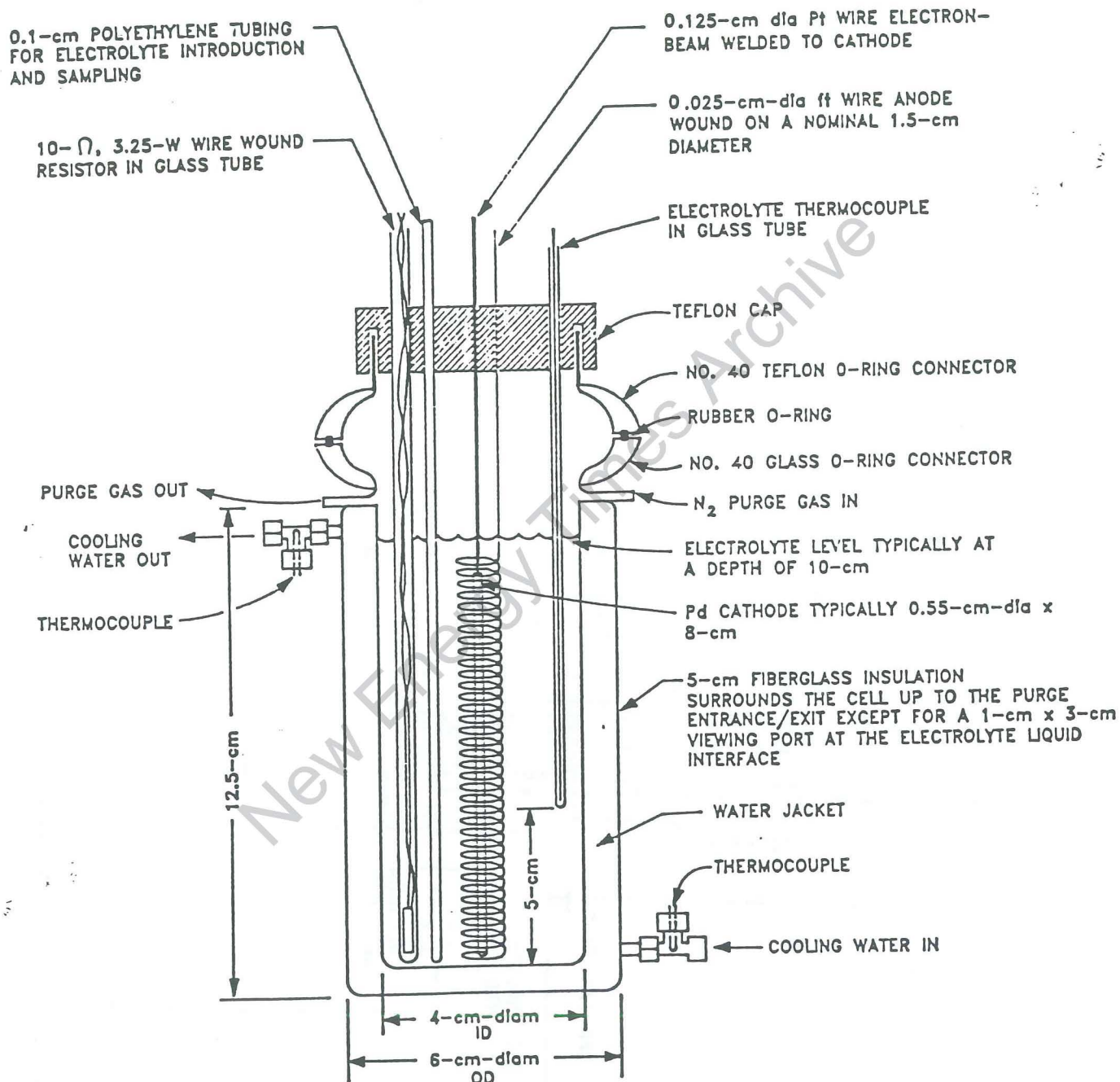
Fig. 3. Top view of the placement of the neutron and gamma-ray monitoring systems.

Fig. 4. Corrected neutron flux as a function of time in test CF-1 with a 0.28-cm diam x 8.5-cm palladium cathode in 0.2 N LiOD operating at 100 mA/cm<sup>2</sup>.

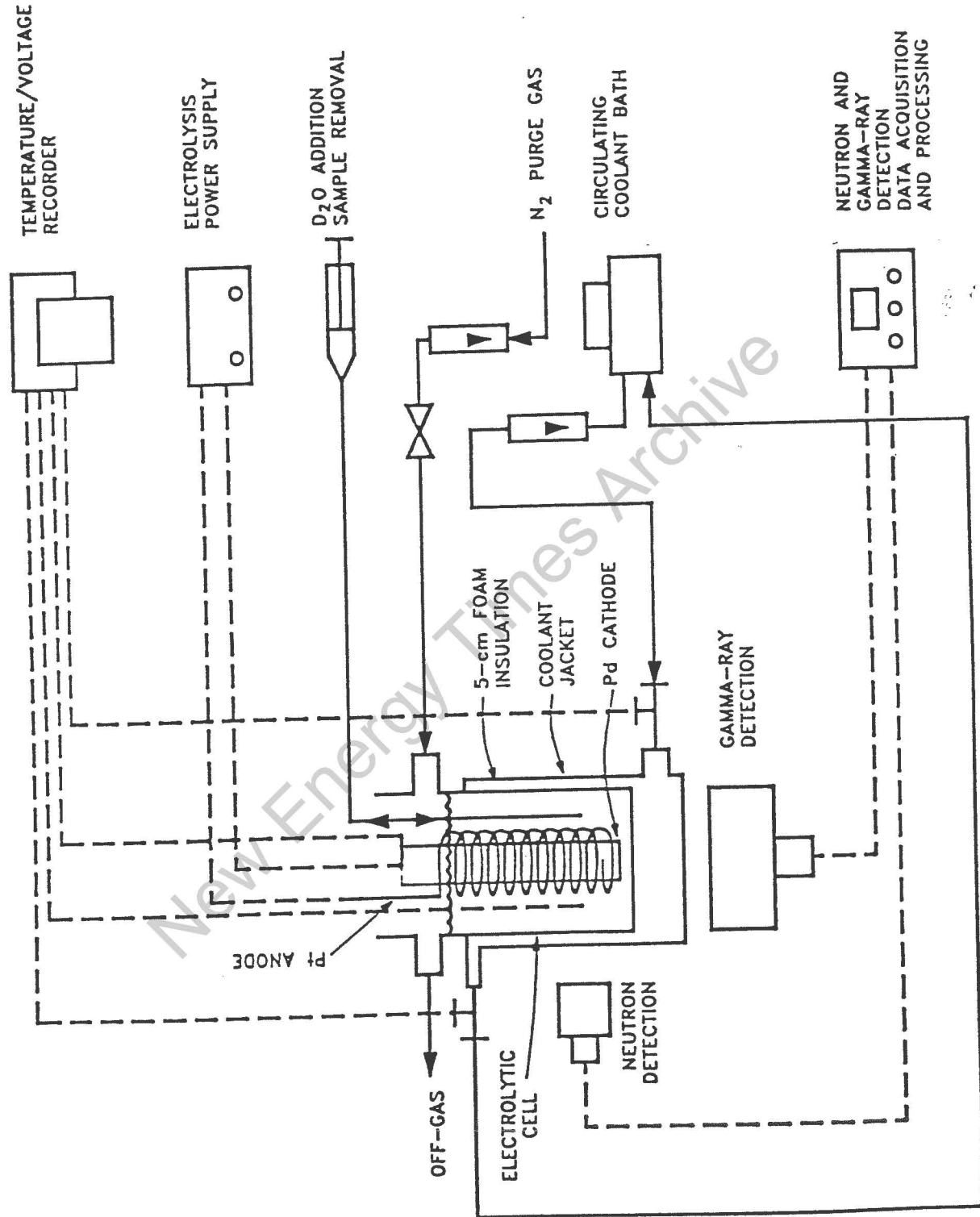
Fig. 5. A frequency histogram of the neutron flux in test CF-1 indicating the anomalous result of 32 neutrons/4 h.

Fig. 6. Energy balance of test CF-2 with a 0.55-cm diam x 8.5-cm palladium cathode in 0.2 N LiOD operating at 100 mA/cm<sup>2</sup>.

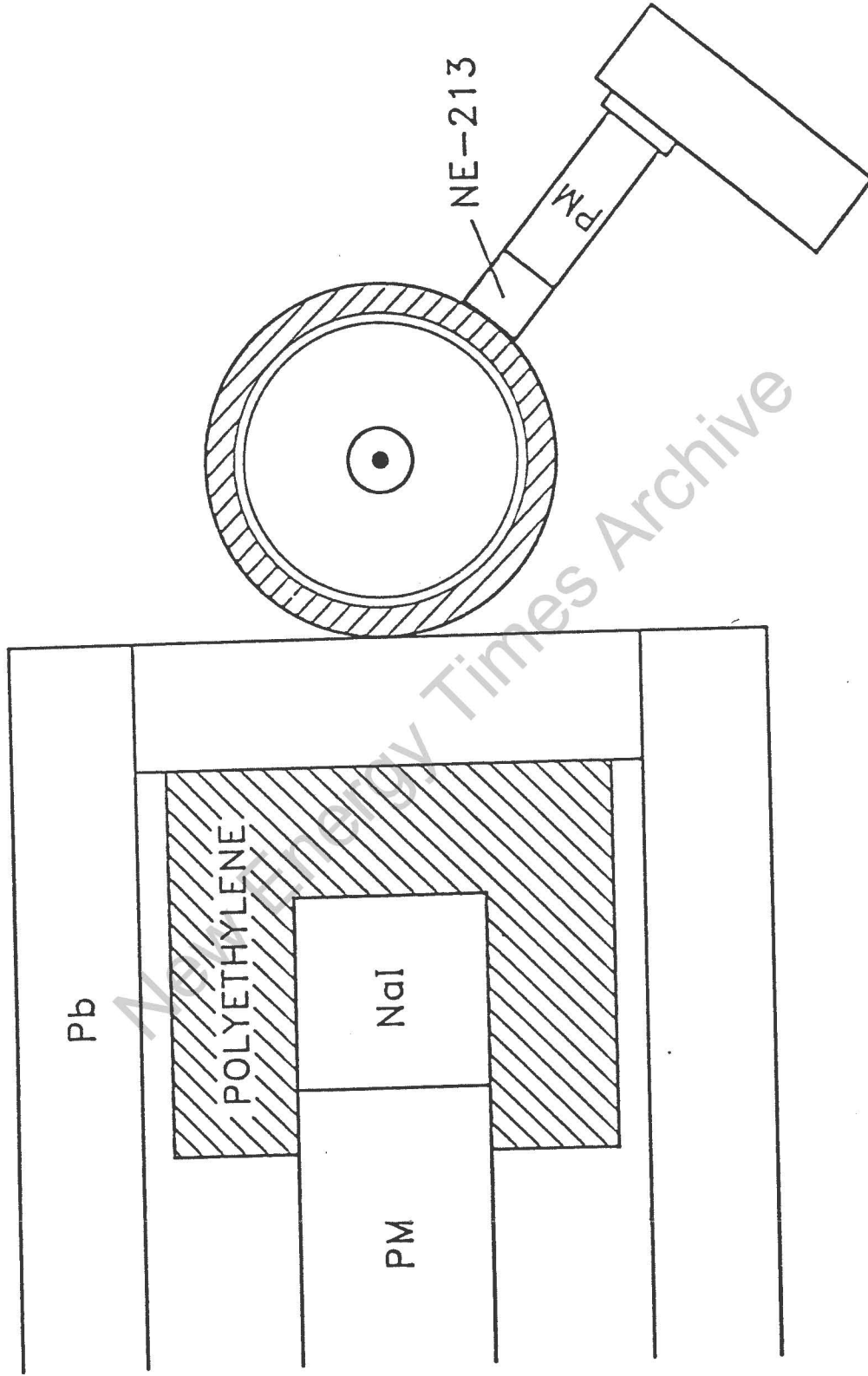
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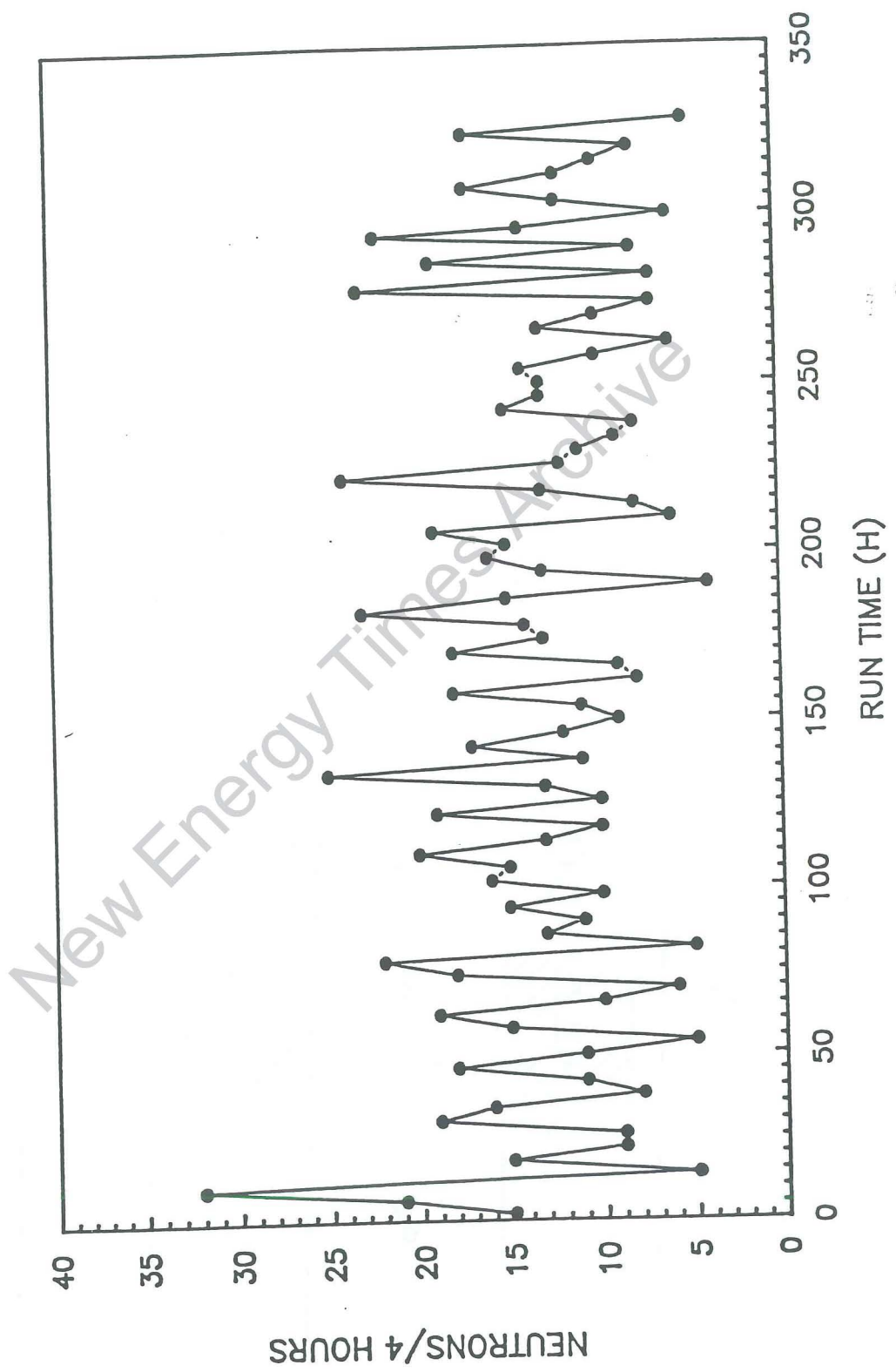
ELECTROLYSIS CELL USED IN COLD FUSION STUDY



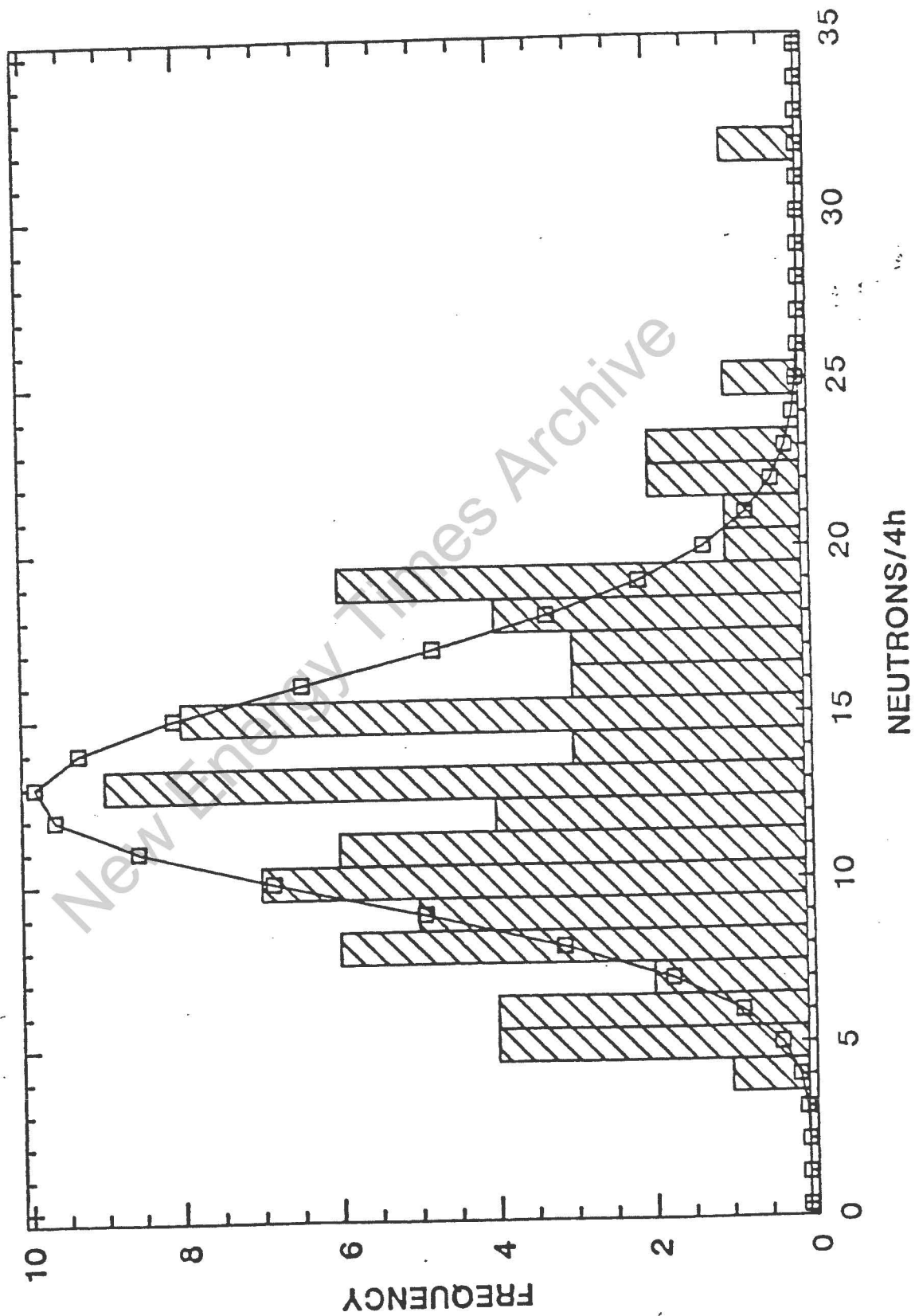
SCHEMATIC OF COLD FUSION EQUIPMENT

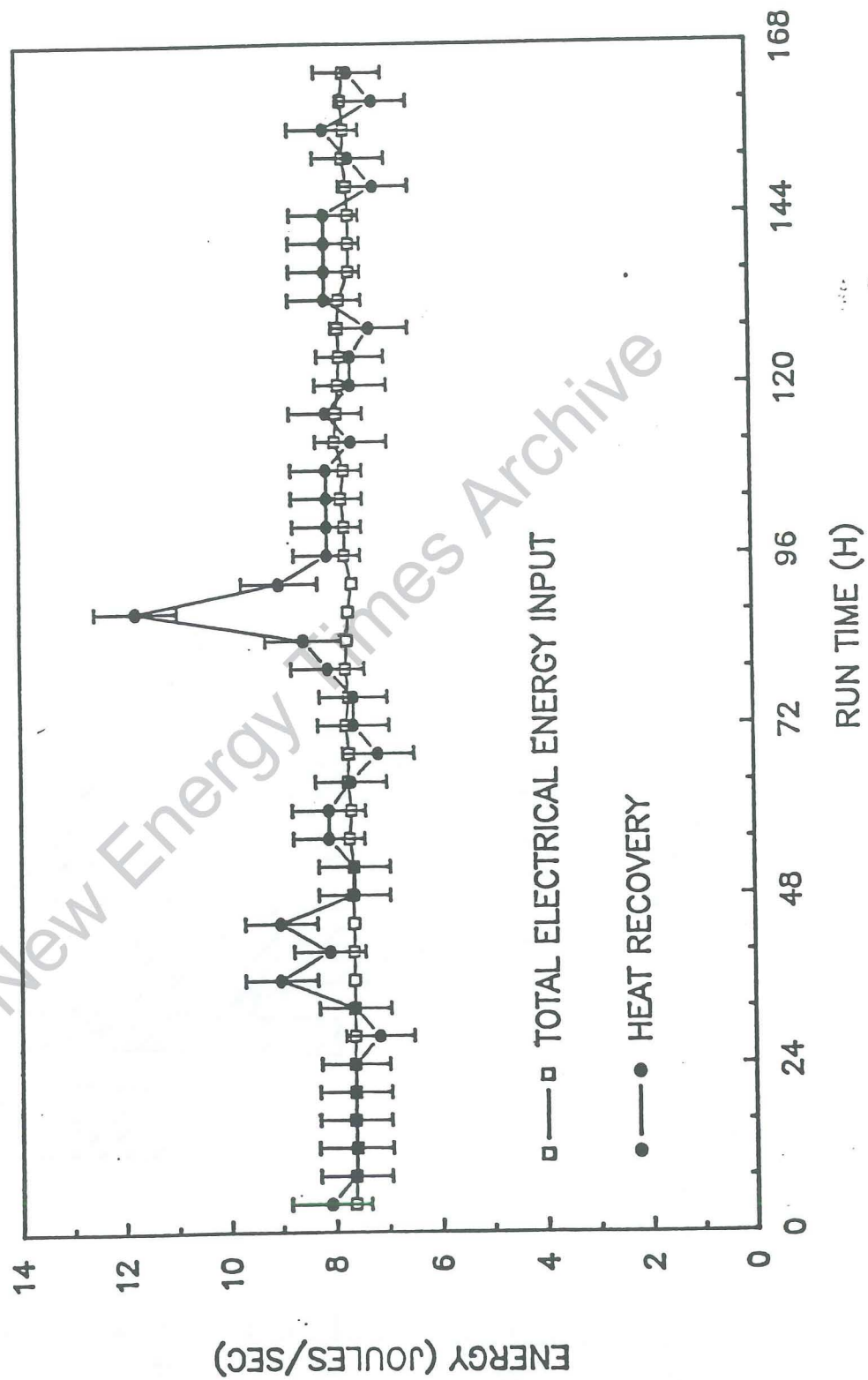


DETECTION SCHEMATIC  
(TOP VIEW)



ORNL DWG 89-524





# **CALORIMETRY EXPERIMENTS IN THE PHYSICS DIVISION--ORNL**

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**ORNL Physics Division**

**J. Bullock IV and G. L. Powell**  
**Y-12 Development Division**

**September 14, 1989**

# CALORIMETRY EXPERIMENTS IN THE PHYSICS DIVISION-ORNL

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## ABSTRACT

Four calorimetry experiments were performed with palladium cathode electrolysis cells to investigate the possibility of cold fusion heat production based on the reported results of Fleischmann and Pons.<sup>1</sup> Two of the cells contained 6.35 mm diam  $\times$  10 cm long palladium cathodes in a 0.2 M Li<sup>6</sup>OD electrolyte, one cell contained a similar cathode in a 0.1 M Li<sup>6</sup>OD electrolyte solution and one cell used a cast 1.27 cm diam  $\times$  10 cm palladium rod in a 0.2 M electrolyte. All four cells were constructed with platinum wire anodes. One of the cells exhibited an apparent 2-3 watt power excess for a period of approximately 300 hours of a total operating time of 1800 hours; each of the remaining cells remained in power balance for the 1800 hour period.

## INTRODUCTION

Immediately following the announcement by Fleischmann and Pons at the University of Utah of the discovery of cold fusion in an electrolysis cell containing a palladium cathode, several experiments were begun at ORNL to investigate this claim. A number of electrolysis cells were constructed using palladium foil cathodes and platinum gauze anodes in an attempt to measure cold fusion produced neutrons. A neutron counter was assembled using NE213 scintillator detectors, which employed pulse shape discrimination

to reduce the effective gamma ray background. The background count rate of this detector was approximately 90 events per hour. Another detector located near the primary system was used to monitor any changes in the background level. No neutrons above the background rate were detected during these initial experiments. We then focused our attention on the report of excess heat result also made by the University of Utah group.

### EXPERIMENTAL APPARATUS

Four calorimetry cells were constructed with an electrolyte volume of 300 cc in a 54 mm o.d. quartz envelope. A diagram of the calorimetry cells is shown in Fig. 1. Each cell contained a teflon plug in the top of the container with a number of penetrations for electrical connections, heavy water refilling, and sampling. Two 6.35 mm teflon rods projected through the top plug to support the palladium cathode rod in the center of the envelope. The teflon rods were spring loaded to allow for expansion of the palladium electrode during charging with deuterium. The anode was helically wound from platinum wire on the inside of a perforated quartz tube. The anode helix was wound on a mandrel and allowed to spring against the inside of the quartz tube for support. No internal recombiner was used to catalyze the recombination of the gases produced by electrolysis back into heavy water. The  $D_2$  and  $O_2$  gases were allowed to escape through a vent hole in the top teflon plug. The quartz cells were placed inside teflon cylinders that in turn were immersed in a temperature controlled water bath. A 2.5 mm gap between the quartz and the teflon cylinder was filled with  $D_2O$  and contained a platinum resistance

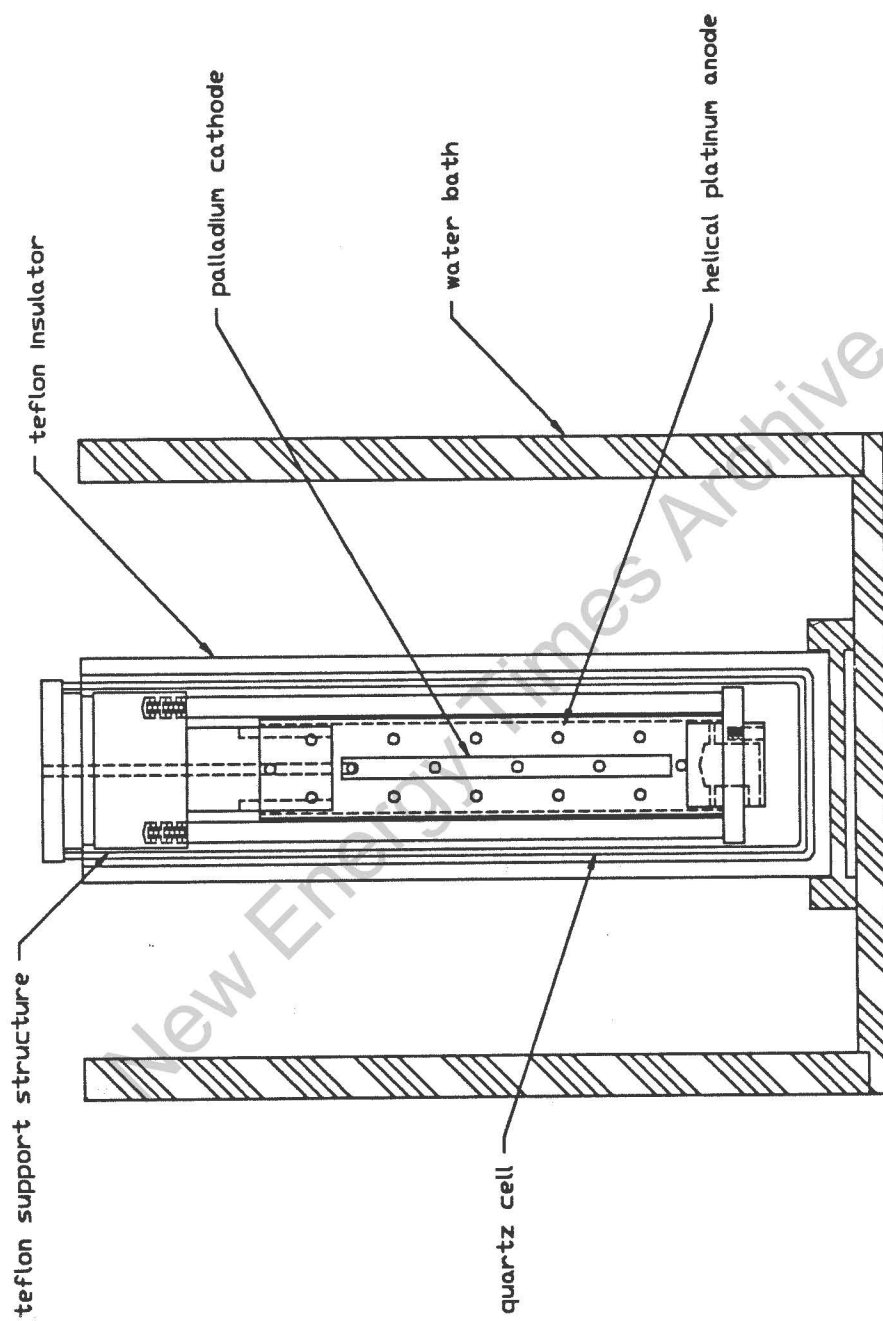


Fig. 1. Diagram of the design of the calorimetry cells.

temperature detector (RTD). Another platinum RTD was placed in the water bath adjacent to the teflon socket. A cell containing a resistance heater similar in size to the palladium cathode and identical in construction to the actual cells was placed into each teflon socket, prior to activation of the cells, for calibration. Varying amounts of electrical power were applied to the resistance heater and the temperature difference across the teflon was noted for each setting. Calibration curves for three of the cells are shown in Fig. 2. The calibration data were fitted with a fourth order polynomial for use with the cell calorimetry data. The second order term was very small and the third and fourth order terms nearly zero. The cells were periodically re-calibrated during the course of the experiments, as well as whenever the bath temperature was altered. A shielded neutron counting chamber was prepared next to the constant temperature baths to permit neutron counting of the cells during operation without interruption of the electrolysis.

### ELECTRODE PREPARATION

Two of the cells contained 6.35 mm diam  $\times$  10 cm long annealed palladium cathodes, one of which was pre-charged in  $D_2$  gas to a stoichiometry of 0.6, in a 0.2 M  $Li^6OD$  electrolyte; one cell contained a pre-charged annealed cathode in a 0.1 M  $Li^6OD$  electrolyte solution; and one cell used a cast 1.27 cm diam  $\times$  10 cm palladium rod in a 0.2 M electrolyte. Annealing was performed by heating the rods to a temperature of 900°C for 4 hours in vacuum. The cast 1.27 cm diam rod and the 6.35 mm diam non-pre-charged rod were baked at 200°C in vacuum for 24 hours to remove any residual protium before being placed in operation. The experimental parameters for the palladium

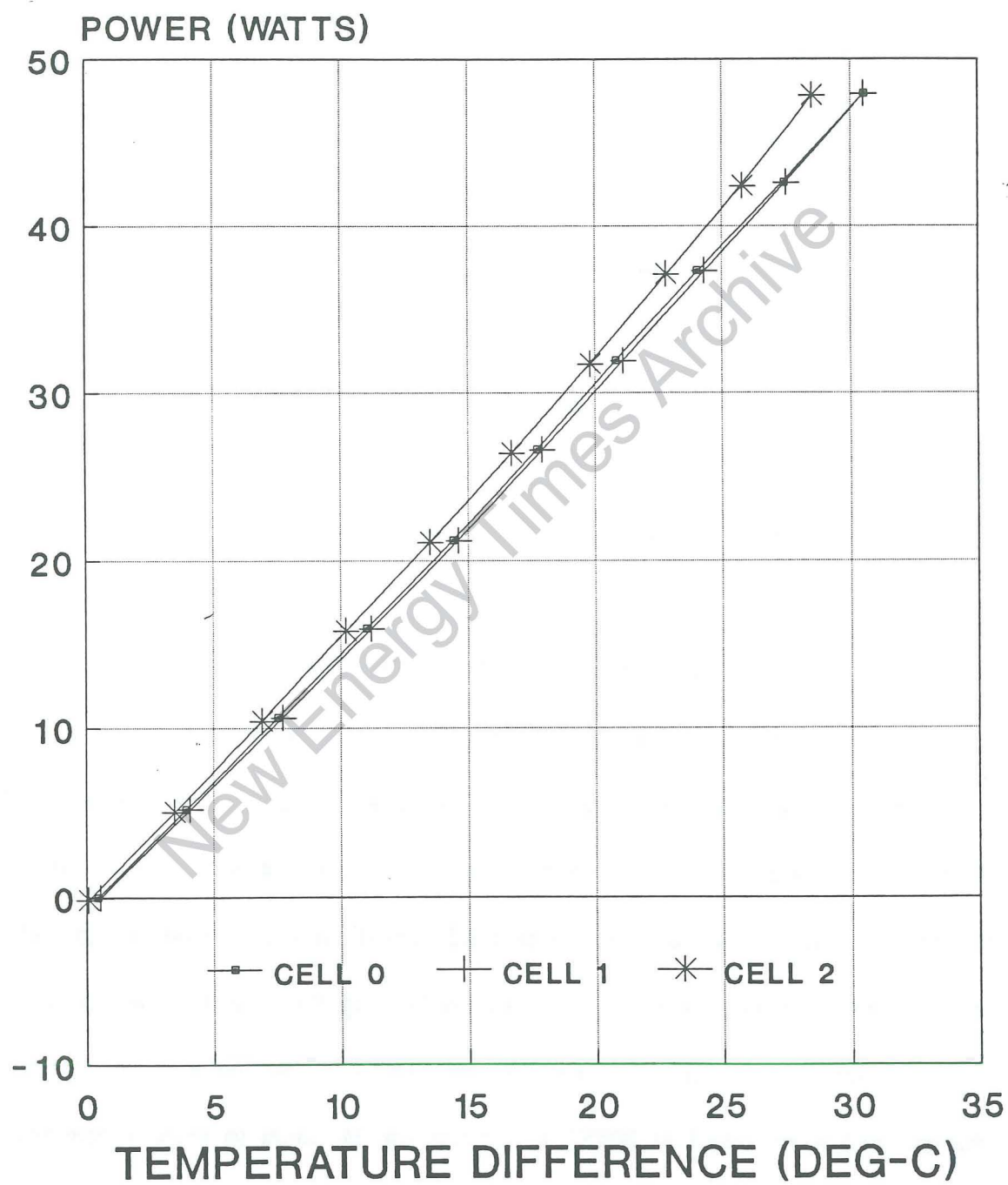


Fig. 2. Calibration curves for three of the calorimetry cells.

cathodes are summarized in Table 1. The three 6.35 mm o.d. rods were purchased from Johnson-Matthey. The cast rod was fabricated from stocks of palladium obtained at ORNL.

## RESULTS

The cells were operated at an initial current density of 50 mA/cm<sup>2</sup> for 48 hours and then at a current density of 250 mA/cm<sup>2</sup> for over 1800 hours. Referring to the cell numbering from Table 1, cells #1, #2, and #3 have remained in power balance within an experimental uncertainty of  $\pm 1$  watt for the duration of the experiments. However, cell #0 exhibited an apparent excess power level of approximately 3 watts for a 300 hour period beginning 180 hours after the beginning of the experiment. This imbalance was approximately 10% of the input power. Our calorimetry cells did not use an internal catalyst to promote the recombination of the electrolyzed gases back into heavy water. D<sub>2</sub>O was added to the cells every twelve hours to replace the water depleted by electrolysis. By carefully recording the water usage rate, we were able to determine that no recombination was occurring in the cells.

The power balance for our cells was determined from the equation

$$P_{\text{imbalance}} = \text{heat output} + \text{electrolysis} - \text{electrical input}.$$

The heat output of the cells was measured by the calibrated platinum RTD's. The electrolysis term was determined by measuring the make-up water for the cells assuming the exhaust gases were 100% humidified. The electrical input was determined by monitoring the voltage and current going to the cells. The electrolysis cells were driven

TABLE 1

## EXPERIMENTAL PARAMETERS

Cell No.	Electrode Configuration	Electrolyte Concentration	Electrode Conditioning
0	6.35 mm diam	0.2 M LiOD	Annealed
1	6.35 mm diam	0.2 M LiOD	Annealed/ pre-charged/ Arsenic added
2	6.35 mm diam	0.1 M LiOD	Annealed/ pre-charged
3	12.7 mm diam	0.2 M LiOD	Cast

**Note:**

1. All rods were 100 mm long.
2. The 12.7 mm diam rod was dropped cast in a chilled copper mold. The rod was x-rayed to identify any possible voids.
3. All rods were heated in vacuum to remove residual protium.
4. Current Density = 250 mA/cm<sup>2</sup>

by computer-controlled constant current sources and the voltages across the electrodes were monitored by a digital-to-analog converter in the control computer. The constant current power supplies were calibrated prior to the experiment. At an operating current density of 250 mA/cm<sup>2</sup>, the total current to the 6.35 mm diam rod cathodes was 5.03 amps. At this current, 1.88 grams of D<sub>2</sub>O were electrolyzed per hour of operation, resulting in volumetric water usage rate of 1.702 cc/hour. Using the value of 70.41 kcal/mole for the heat of formation of heavy water, this electrolysis rate results in 7.635 watts of power consumption. Assuming that the exhaust gases have a relative humidity of 100%, the total water consumption rate we calculate is 1.88 cc/hour corresponding to a total electrolysis power level of 7.77 watts, including power due to evaporation. The consumption of D<sub>2</sub>O was monitored over the entire course of the experiment. Figure 3 shows a graph of the measured replenishment rate for the first 21 days of the experiment. The open circles are the measured values and the straight line is a straight line fit to the data points. The fitted value of 1.87 cc/hour is in excellent agreement with the predicted value of 1.88 cc/hour. Using this result the heat balance equation becomes

$$P_{\text{imbalance}} = \text{heat} + 7.77 \text{ watts} - V_{\text{cell}} \times I_{\text{cell}}$$

In addition to control of the power supplies and constant monitoring of the cell parameters, the control computer was programmed to sweep the cell current once each hour. The duration of the current sweep was 5 seconds. The slope of the I-V curve is the electrolyte resistance.<sup>2</sup> By multiplying the square of the cell current by this quantity, the ohmic power deposited in the electrolyte may be determined. Figure 4 contains a plot of the measured parameters of cell #0 for the first 600 hours of operation. The lower

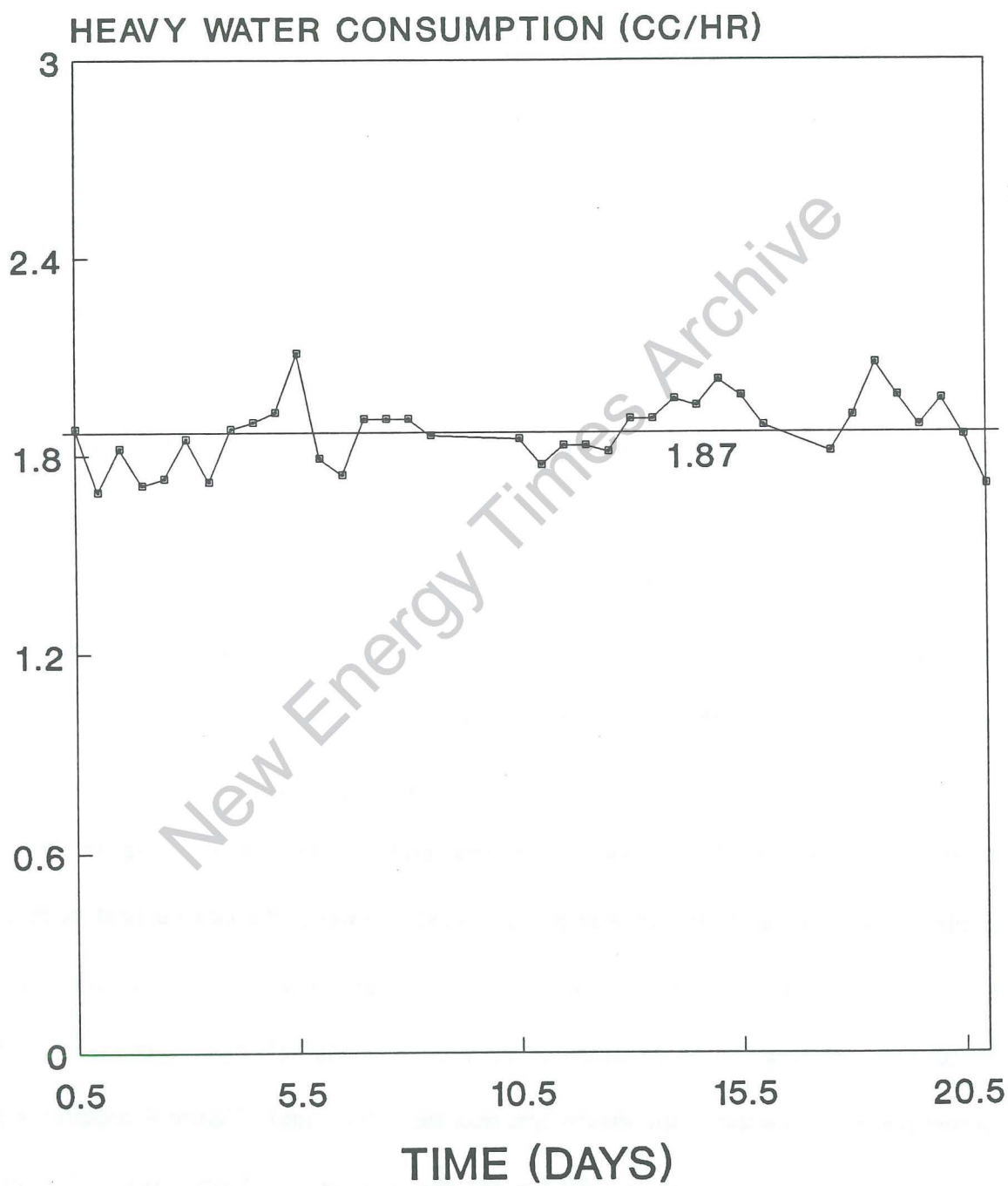


Fig. 3. Heavy water consumption in cell #0 for the first 21 days of operation.

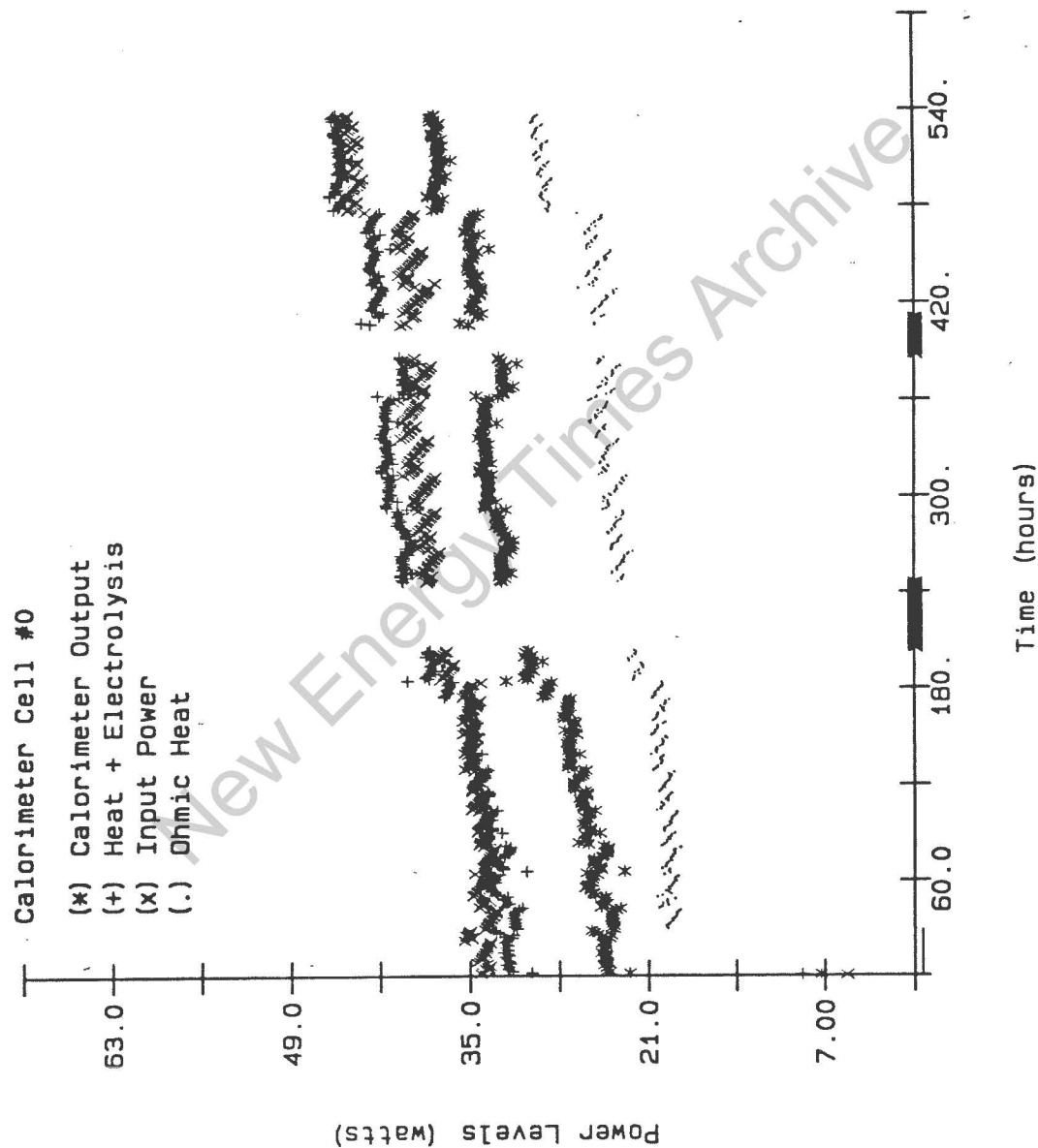


Fig. 4. The measured output parameters of cell #0.

curve, represented by the dots, is the ohmic power,  $I_{\text{cell}}^2 \times R_{\text{electrolyte}}$ , deposited in the electrolyte. The curve represented by the asterisks is the measured calorimeter power output determined by comparing the temperature difference across the teflon with the calibration curve obtained with the electrical resistance heater. The curve represented by the x's is the electrical input power to the cell,  $V_{\text{cell}} \times I_{\text{cell}}$ . Finally, the curve represented by the (+) symbol is the sum of calorimeter plus the electrolysis power value of 7.77 watts. During the initial 75 hours of the experiment, cell #0 exhibited a slight negative heat balance. At a level of less than 2 watts tapering to zero at approximately 75 hours, this indicates an energy storage of approximately 270 kJoules. This far exceeds the hydride heat of formation value of approximately 3 kJoules calculated for these rods. Cell #0 remained in balance until the 180 hour mark when a positive imbalance of two watts was observed. Twelve hours after this apparent excess heat event began, the temperature of the water bath was inadvertently changed from an initial value of 18°C to 13°. The ohmic power increased at this point due to the increased electrolyte resistance at the lower temperature. The input power also increased with the increased electrolyte resistance because the cells were driven by constant current sources. The calorimeter output also increased with the lower temperature and maintained the 2 watt excess. The sawtooth behavior, which is most evident on the input power and ohmic power curves, with a period of approximately 12 hours occurred because of the changing level of liquid in the cell due to electrolysis and the periodic refilling twice a day. As the liquid level decreases, the heat transfer area changes and the electrolyte resistance decreases as the molality increases. Although the affect of these changes is not accurately known, we only use data taken at

the peaks of the sawtooth, where we know the calibration is accurate, for evaluation. The gap in the data represents a period when the cell was removed from the calorimeter socket for 30 hours. During this period, the socket was re-calibrated at the lower temperature and the cell was placed in the neutron counter. All of the parameters of the cell were monitored during this time except heat output. We arbitrarily chose to display data taken only when the cell was in the socket. The imbalance increased slightly until the 370 hour mark when the temperature of the bath was deliberately raised to 18°C. The imbalance nearly disappeared but recovered and increased over the next 120 hours to slightly over 3 watts. At approximately 480 hours the cell bath temperature was lowered to 5°C in an attempt to study the effect of temperature changes on the imbalance. The excess power disappeared at this point and repeated temperature and current cycling failed to reproduce the effect. The power balance for cell #0 for the first 25 days of operation is shown in Fig. 5. The data were averaged over a twelve hour period for this graph. We continued the experiment for a total period of 1800 hours with no recurrence of the excess heat observation. The other cells remained in accurate power balance during the entire experimental period. During the observation of heat imbalance in cell #0, cell's #0 and #1 were switched in their calorimeter sockets to verify that the apparatus was working properly. The power readings for each cell during this period were consistent with the calibrations for both cells, the measured imbalance in cell #0 remained irrespective of the calorimeter socket used.

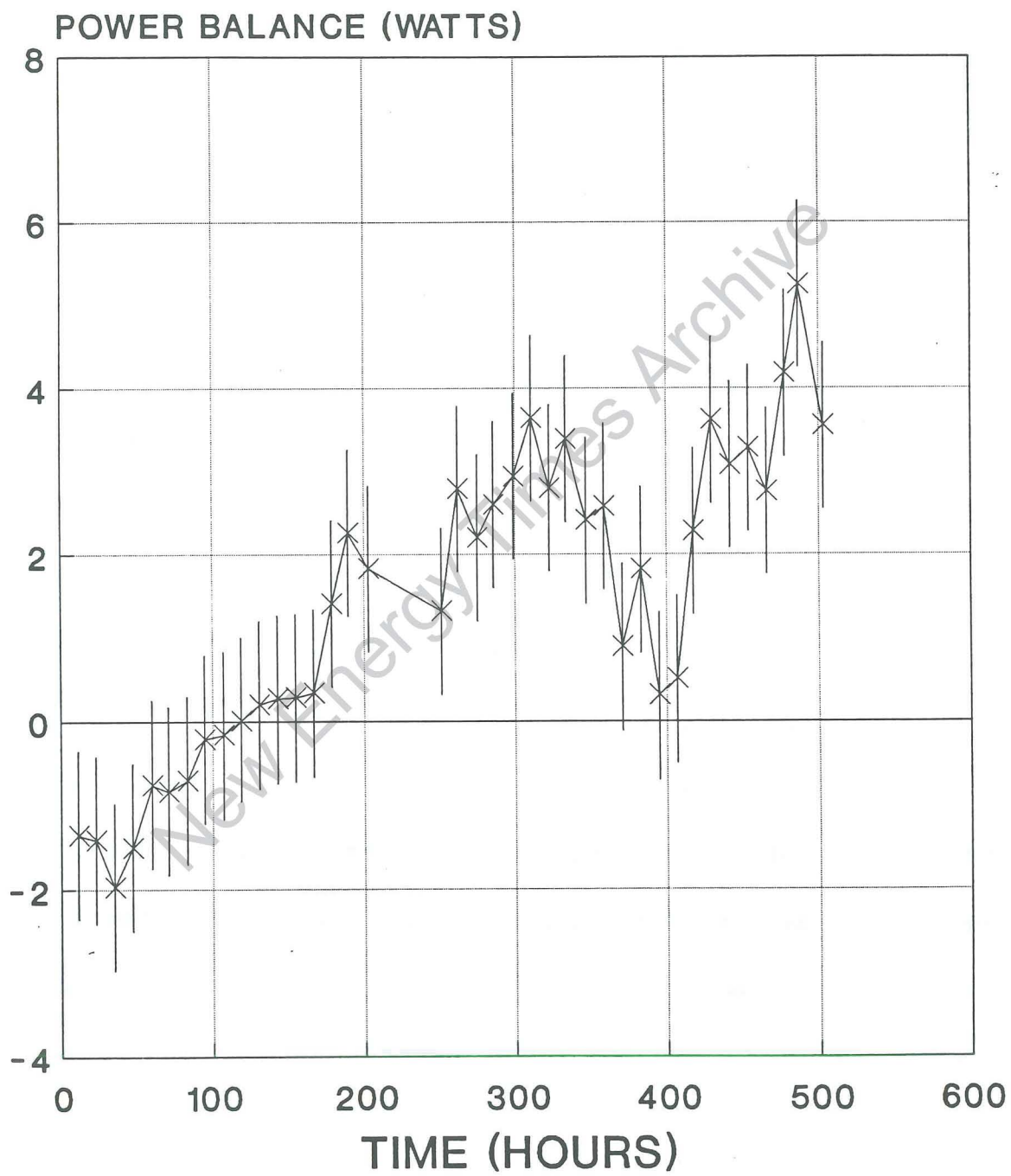


Fig. 5. Power balance plot for cell #0.

## ADDITIONAL MEASUREMENTS

Shortly after the power imbalance was discovered, cell #0 was removed from the calorimetry socket and placed in a shielded neutron counter containing a pair of NE213 scintillator detectors with pulse shape discrimination. The neutron emission level of the cell was determined to be less than  $1 \times 10^{-24}$  neutrons/sec/d-d pair, as set by the background level of the counting system. The tritium level of the initial electrolyte fill of D<sub>2</sub>O was measured to be  $1.0 \times 10^6$  dpm/cc and increased to a level of  $1.3 \times 10^6$  dpm/cc 50 hours after the beginning of the apparent excess power production. The accuracy of the tritium measurement was at best 10 per cent and because of this uncertainty we draw no conclusions from our measurement.

## CONCLUSIONS

During a period of approximately 300 hours, one cell of four exhibited a positive power imbalance of 2-3 watts after a short period of negative imbalance. The mechanism responsible for this behavior has not been identified; it appears to be a transitory effect with some dependence on temperature.

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Fusion Phenomena, Sant Fe, New Mexico, May 23-25, 1989, Martin Marietta Energy  
Systems, Inc., Oak Ridge Y-12 Plant, May 1989.

New Energy Times Archive

## A PRELIMINARY INVESTIGATION OF COLD FUSION BY ELECTROLYSIS OF HEAVY WATER

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### SUMMARY OF RESEARCH FROM APRIL 1 TO AUGUST 31, 1989

The initial goal of this research has been to experimentally study possible thermal and nuclear reactions occurring during the electrolysis of  $D_2O$  with a  $LiOD$  electrolyte and a palladium cathode.

#### System Description

Insulated Pyrex glass electrolysis cells with active internal geometries of  $\sim 4\text{-cm-diam} \times 10\text{ cm}$  were used with palladium cathode rods of  $0.28\text{- to }0.55\text{ cm-diam} \times 8\text{- to }9\text{-cm}$  operating at  $100\text{ to }600\text{ mA/cm}^2$ . Heat removal was determined by the temperature increase of the forced flow of cooling water. Calibrated thermocouples allowed temperature measurements to  $\pm 0.05^\circ\text{C}$  and the coolant water pumping rate was controlled with an accuracy of  $0.5\%$ . Typically this resulted in an error bar of  $\pm 0.2$  watts in the recovered heat or  $2\text{ to }5\%$  of the total energy input.

Most of the tests have been made with "open" systems in which the electrolysis gases were allowed to freely escape from the cell. However, in the most recent test, the electrochemical cell has been designed with additional internal vapor space that enclosed a catalytic system for recombination of the  $D_2$  and  $O_2$ . This system consists of a Pt wire coil electroplated with Pd black which can be heated by imposition of electrical current. The recombiner can quantitatively combine the  $D_2$  and  $O_2$  electrolytically produced by currents as high as  $2.13\text{ A}$  (typically, a current density at the cathode of  $300\text{ mA/cm}^2$ ). This "closed" system actually operates with a slight vacuum and there is no required addition of  $D_2O$  or electrolyte except for replacing the electrolyte samples taken for analysis.

Neutron count rates were measured by a NE-213 scintillator with pulse-shape discrimination and a threshold of 1.2 MEV that had a typical sensitivity at three standard deviations of  $3 \times 10^{-24}$  fusions/ deuterium-pair-s. A gamma-ray spectrometer utilizing NaI was used for detection of high-energy gamma rays, but it had somewhat less sensitivity due to the higher gamma-ray background. The tritium concentration was measured periodically in the electrolyte.

Data acquisition was by two on-line computer systems, one for recording the gamma-rays and neutron count rates, and the other for the recording of the temperatures, voltages and coolant water flow rate. A back-up strip-chart recorder was also used for this purpose.

### Experimental Results

With the assumption that there was no recombination of  $D_2$  and  $O_2$  within the cell, heat in excess of that provided by joule heating (IxE) was detected for periods of many hours in some of the open-system experiments. This was usually in the range of 5 to 10%; however, during one 12-h period, excess power of up to 50% was seen. The experimental uncertainty was calculated to be 3 to 5%. The periods of excess power, which were transitory, coincided with increases in current density. They could be extended by perturbing the system, for example changing the electrolyte temperature or increasing the electrolyte concentration.

During the first 740 h of operation with the closed system, the power balance was slightly negative (typically 1 to 2%), indicating a slight heat loss through the insulation of the cell. Starting at 740 h and continuing after more than 60 additional h there has been an increasing positive power balance that is now in excess of 5%. The estimated error of measurement is approximately  $\pm 2\%$ .

There have been two separate occasions within the first 90 h of operation with two different 0.28-cm-diam Pd electrodes when the neutron count rate exceeded three standard deviations above the background measured prior to the start of the experiment. These higher neutron count rates were uncorrelated with any excess power observations. In the present closed-system test, there has also been a neutron count rate that exceeds three standard deviations, but in this case it has occurred during a time of increasing excess power.

During one test, tritium concentration in the electrolyte increased by at least a factor of 10 during the first two days of the experiment. No further increases were observed during the remainder of that experiment. Again, this was an isolated incident which did not coincide with observed periods of excess heat or when neutron count rates exceeded three standard deviations above background.

### Future Plans

These preliminary results have not been reproducible and they are not sufficiently definitive to confirm known cold fusion mechanisms. However, the apparent unusual phenomena are extremely interesting and will be basis for additional investigation. The research in Chemical Technology Division will continue with an emphasis on a system design that will allow a more controlled environment so that nuclear events or excess energy can be measured with more assurance. A closed-system approach with internal recombination will continue and a future system will utilize a continuously flowing electrolyte solution that is also the heat exchange medium. This design concept represents an appropriate processing approach for scale-up should the phenomena prove useful and controllable.

Of particular interest, is the determination of the system parameters that tend to initiate energy excess or nuclear events. These include chemical and physical characteristics and the use of process instabilities such as temperature or electrical current variation. The chemical and physical properties of the electrodes will be carefully determined, and the chemical characteristics of the electrolyte solutions including tritium concentration will be more carefully monitored. Electrodes that are associated with extended positive results will be further investigated for chemical content including deuterium, lithium isotopes, tritium and helium and for physical variations such as crystalline structure, micro cracks, etc. Other electrode materials and electrolyte constituents will be investigated in future tests.

## Plasma Physics Laboratory

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Subject: Summary Report of PPPL Cold Fusion Experiments to the ERAB,  
pursuant to W. L. Woodard to H. F. Furth, 5 Jun 1989,  
pursuant to Watkins to Schoettler, 24 Apr 1989

Filed by: D. M. Manos, Head, PPPL Ad-hoc Task Group on Cold Fusion

Motivated by press releases on cold fusion, PPPL, in association with Electron Transfer Technologies, Inc. of Princeton, NJ, undertook a modest experimental program to investigate certain claims. At PPPL, two apparatus were set up, one to count neutrons, one to count gamma rays.

### APPARATUS

A schematic drawing of the neutron counting apparatus is shown in Figure 1 and a block diagram of the counting circuitry employed for this apparatus is shown in Figure 2. Not shown in Figure 1 is an interior lid of cadmium absorber material which was present on top of the inner steel drum. The apparatus is capable of displaying the neutron energy spectrum in addition to the counts which fall within a selected range of energies accumulated during adjustable periods of time. The range was adjusted so that only events corresponding to the deposition of 2 to 3 MeV in the  $\text{BF}_3$  detectors would register on the counter. The device was calibrated frequently using a  $^{252}\text{Cf}$  source. The background neutron rate was highly variable, ranging from 3 to 30 counts/hr. The neutron efficiency was 2% for experiments using the central test cell, 0.3% for most experiments using the outer test cell, and 0.1% for certain few experiments which used the outer test cell and a modified arrangement of shielding materials. Figure 3 shows a comparison of a background spectrum and the spectrum of the  $^{252}\text{Cf}$  source. We claim that our apparatus can continuously resolve a neutron source strength of 1 n/s.

Figure 4 shows a schematic, including a block diagram of the counting equipment, for the gamma ray counting apparatus. The counter, borrowed from the Colorado school of Mines, consists of a 4" dia. by 4" long, cylindrical NaI crystal, surrounded by a 4 inch i.d., 12" o.d. by 8" long, annular NaI crystal. The interior crystal is fitted with a single photomultiplier, the annular crystal is fitted with 6 photomultipliers. The geometric arrangement is shown in Figure 4. When a gamma ray transits the annular NaI crystal the resulting signal from one of the 6 outer photomultipliers is used to gate the counting electronics so as not to count the signal from the interior photomultiplier should that gamma ray also transit the interior crystal. This provides a very effective discrimination against the cosmic ray background. This apparatus also permits display of the energy spectrum along with the total counts within a selected energy range during an adjustable counting time interval. The energy range selected was 5-6 MeV, in an attempt to observe the 5.4 MeV gamma production associated with the p-D fusion reaction. The detection efficiency for the apparatus is 0.3 %. The observed background count rate with the anti-coincidence veto active was 10 to 25 counts/hr., the background with the veto off was 275 counts/hr. Figure 5 compares of backgrounds with the veto on and off, indicating the observed spectrum in our laboratory, and showing the selected energy window. We claim that this apparatus can continuously resolve a gamma source strength of 1.5 photons/s.

## EXPERIMENTS

Using these counting apparatus we studied the following different types of "cold fusion" systems:

- 1) Electrochemical cell using Pd rods and spheres, metallurgically prepared both by casting and drawing using LiOD in D<sub>2</sub>O at current densities up to 0.6 amps/cm<sup>2</sup>. A similar cell was studied at Electron Transfer Technologies to determine whether energy and power balance were achieved.
- 2) Electrochemical reactor using Pd wires and spheres, metallurgically prepared by casting and drawing using LiOH in 50% H<sub>2</sub>O/50% D<sub>2</sub>O. (In addition to the D-D reaction, the p-D reaction was chosen for careful study to test the hypothesis that fusion may occur by tunnelling through a coulomb barrier at low temperature, mediated by unknown effects from the crystal lattice. Such studies are made easier by the relatively rare occurrence of 5.4 MeV events in the cosmic ray background.)
- 3) Thermal and pressure cycling of Ti turnings as prescribed by workers at Frascati.
- 4) Thermal and pressure cycling of ultra-pure, 200 mesh Ti powder as prescribed by workers at Genoa.
- 5) Numerous variations of items 3 and 4 changing the schedule of variation of pressure and temperature with time.
- 6) Thermal and pressure cycling of Nb bar stock and Nb turnings using the pressure and temperature schedule of Frascati.
- 7) Thermal and pressure cycling of alumina, quartz, and feldspar mixtures (known to have large piezoelectric coefficients and/or to produce persistent high-energy exo-electron emission on fracture).
- 8) Tensile and compressive strain and fracture of Ti sheets loaded with D<sub>2</sub> by glow discharge plasma bombardment.
- 9) Rapid compressive fracture (hammer blows) of Zr/Al/V pellets loaded with D<sub>2</sub> by glow discharge plasma bombardment (after the method of Klyuev et al).

## RESULTS

In every case we saw no neutrons or gamma rays above the measured background, as indicated in the following table:

Experiment	Claimed Level	Observed at PPL
Electrolysis D-D	$4 \times 10^4$ n/s	<1 n/s
Electrolysis p-D	$4 \times 10^4$ photons/s Theoretical	<3 photons/s
Pressure/Temperature Cycling (Frascati, Genoa)	$5 \times 10^3$ n/s	<1 n/s
Fracture studies	3 x background	background
(Klyuev and variants)	(0.4 n/shot)	background
Jones et al	0.4 n/s	not attempted (special electrolyte)

In the electrolytic cell experiments at Electron Transfer Technologies, performed by William Ayres, we found energy and power balance to within +/- 10% for currents up to 0.6 amps/cm<sup>2</sup>. The uncertainties at low currents make the error bars larger than at high current. Figure 6 summarizes our power balance experiments.

## PRESENT STATUS

Based on our own results and those reported by other groups we feel that the proof of the existence of cold fusion now requires unequivocal demonstration to qualified observers by those who claim to be able to detect it. Therefore we have reduced our level of activity in this area. We have returned the gamma ray detector to Colorado School of Mines and have suspended indefinitely, work on electrochemical cells. We have suspended attempts to reproduce pressure and thermal cycling results of the type claimed by Frascati and Genoa pending further reports from the group of Menlove et al, at Los Alamos. We are sustaining a low-priority effort related to further studies of photons and, radio waves, which may result from particle acceleration in electric fields associated with the fracture of non-conductors and metals. These studies are being conducted in the surface physics branch by an incoming graduate student.

cc: T. Brolin  
H. F. Furth  
L. Ianello, DoE  
Members ad-hoc cold-fusion group  
PPPL Division Heads

#89X0302

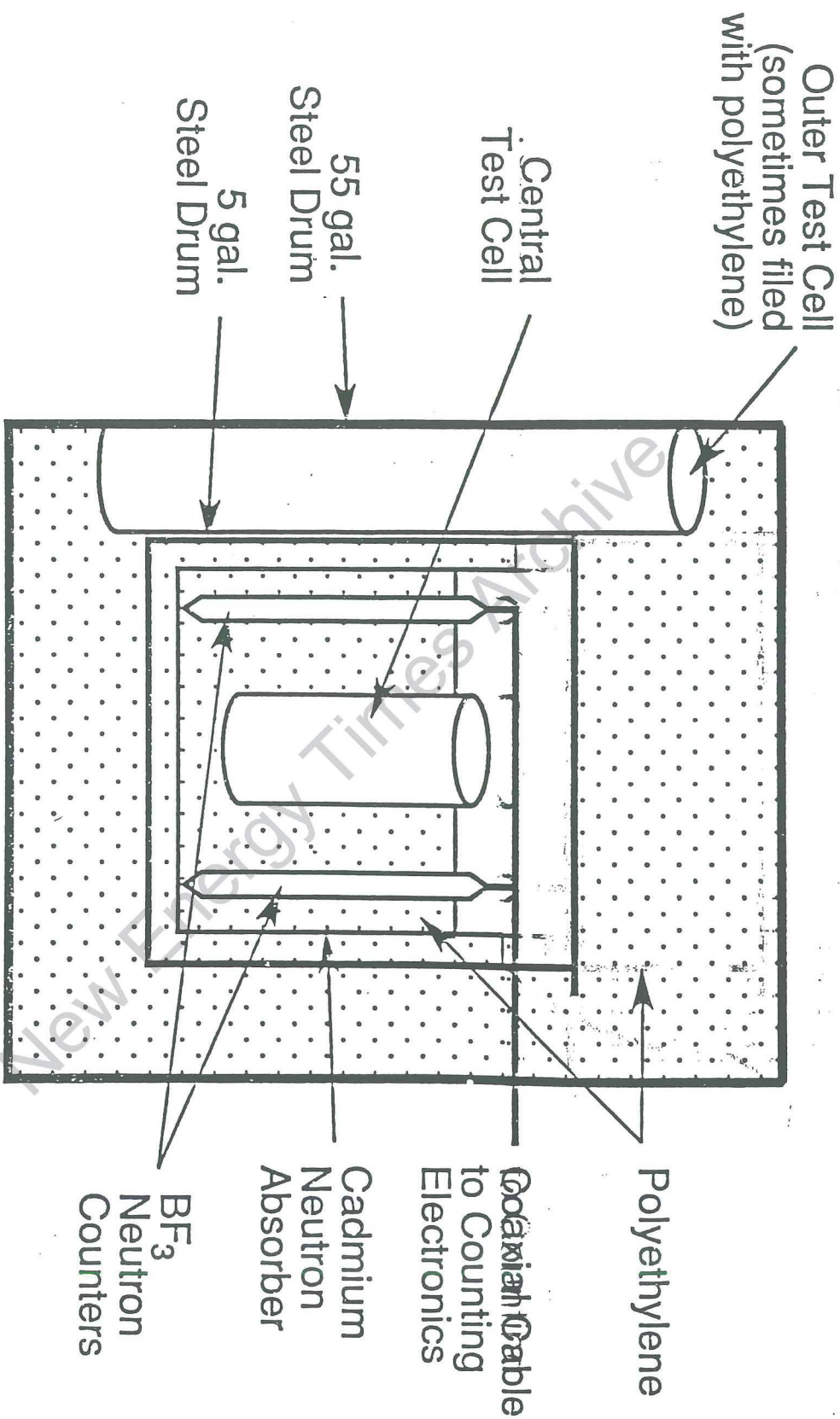


Figure 1

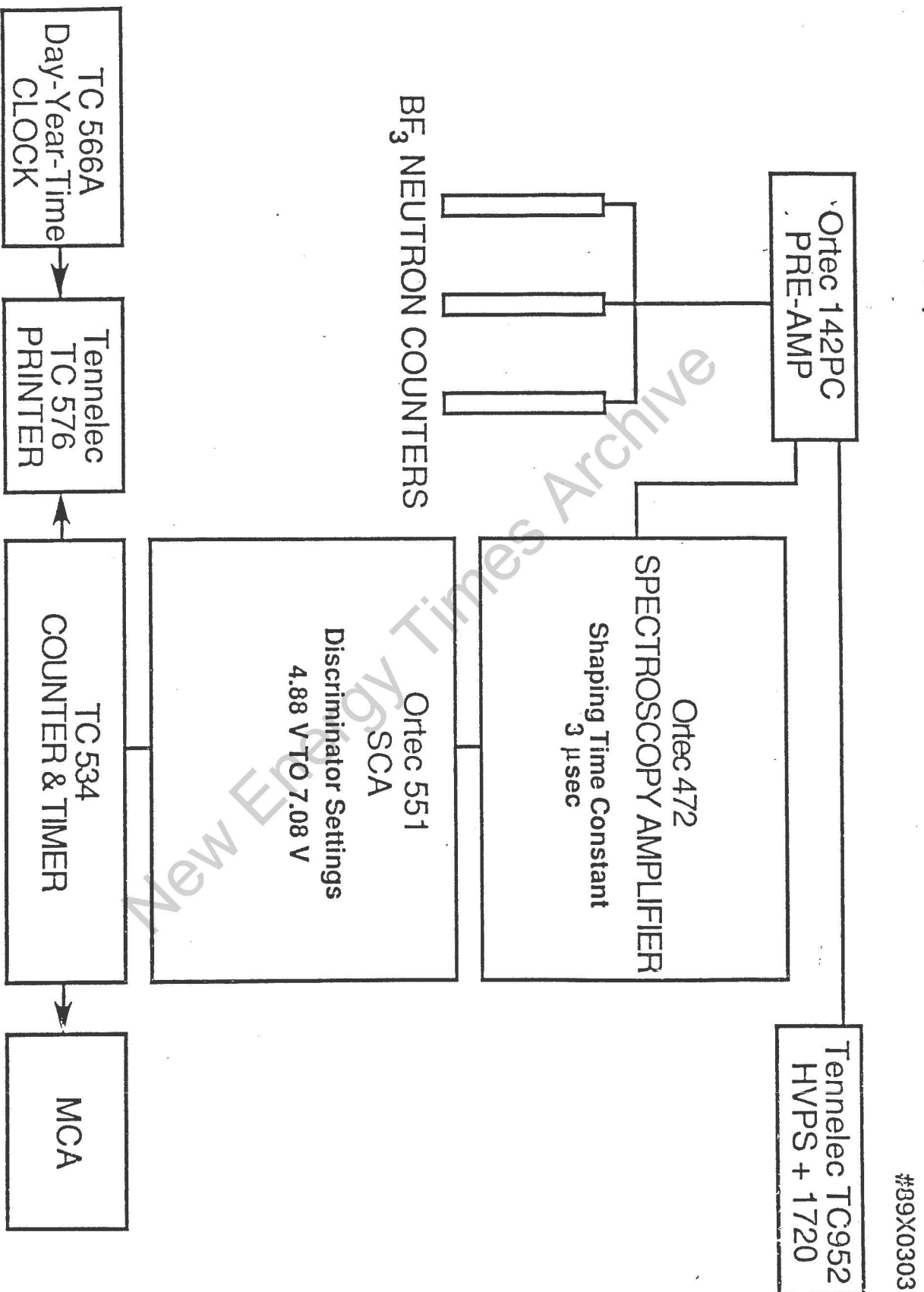


Figure 2

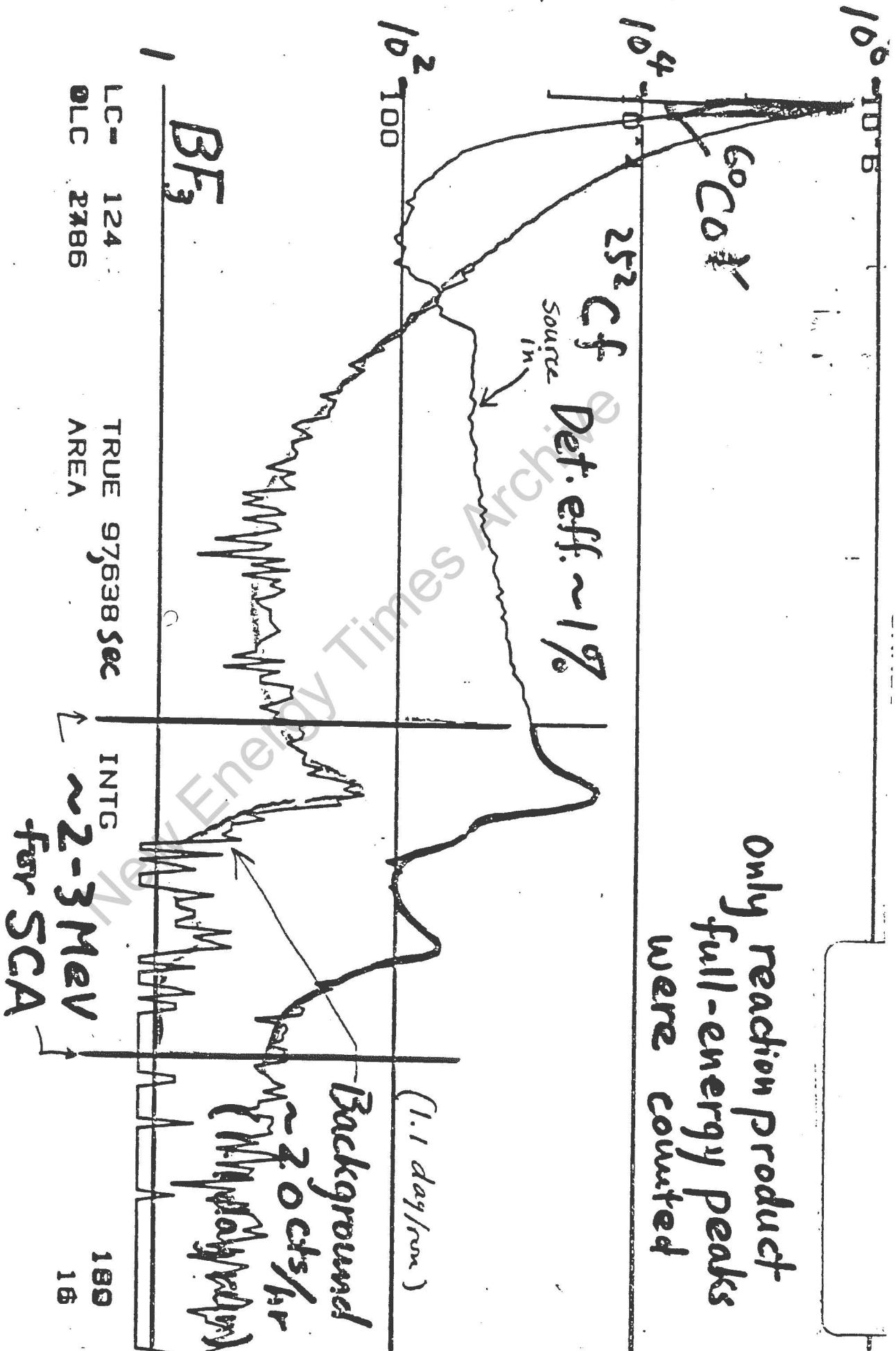


Figure 3

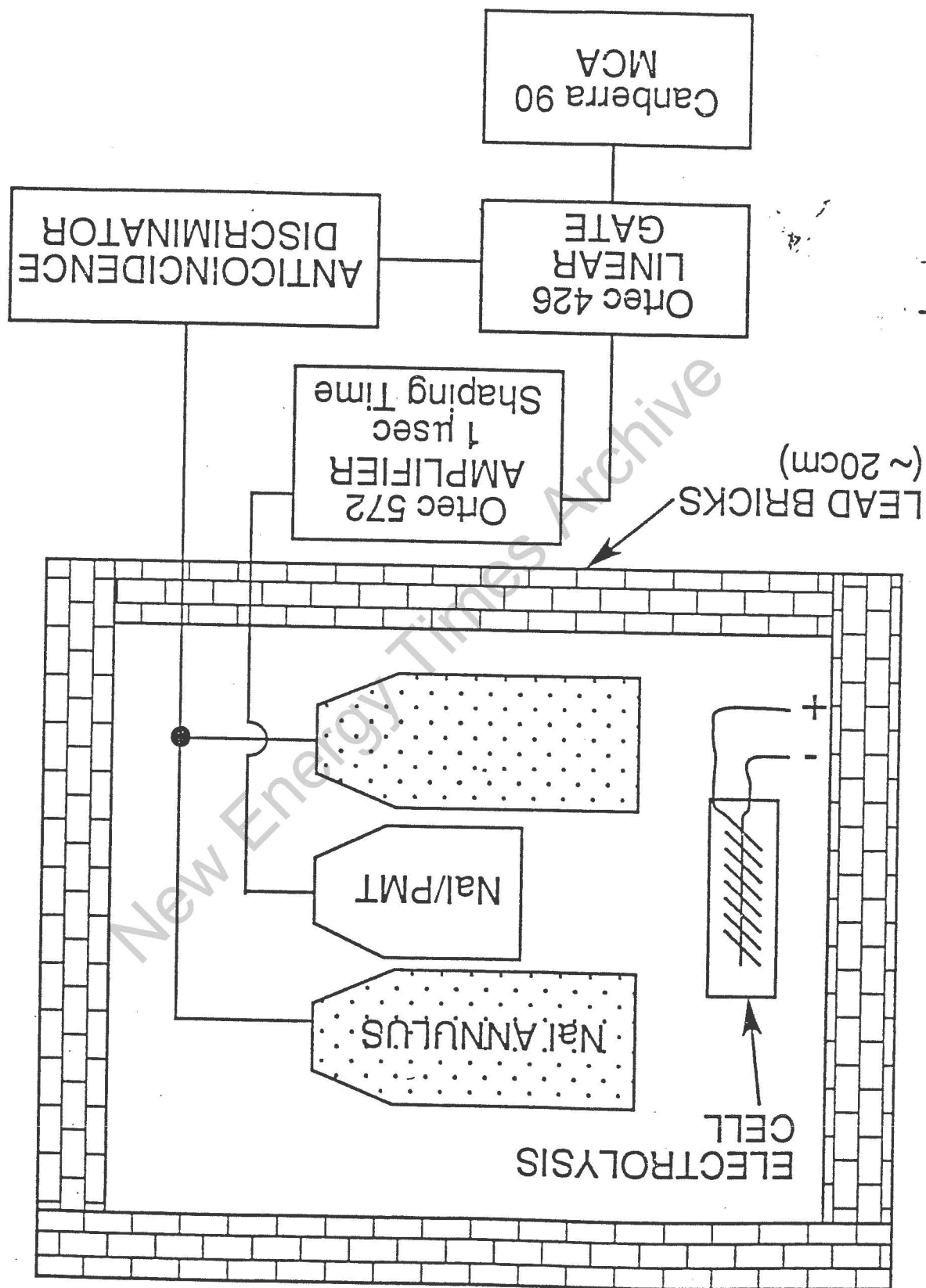


Figure 4

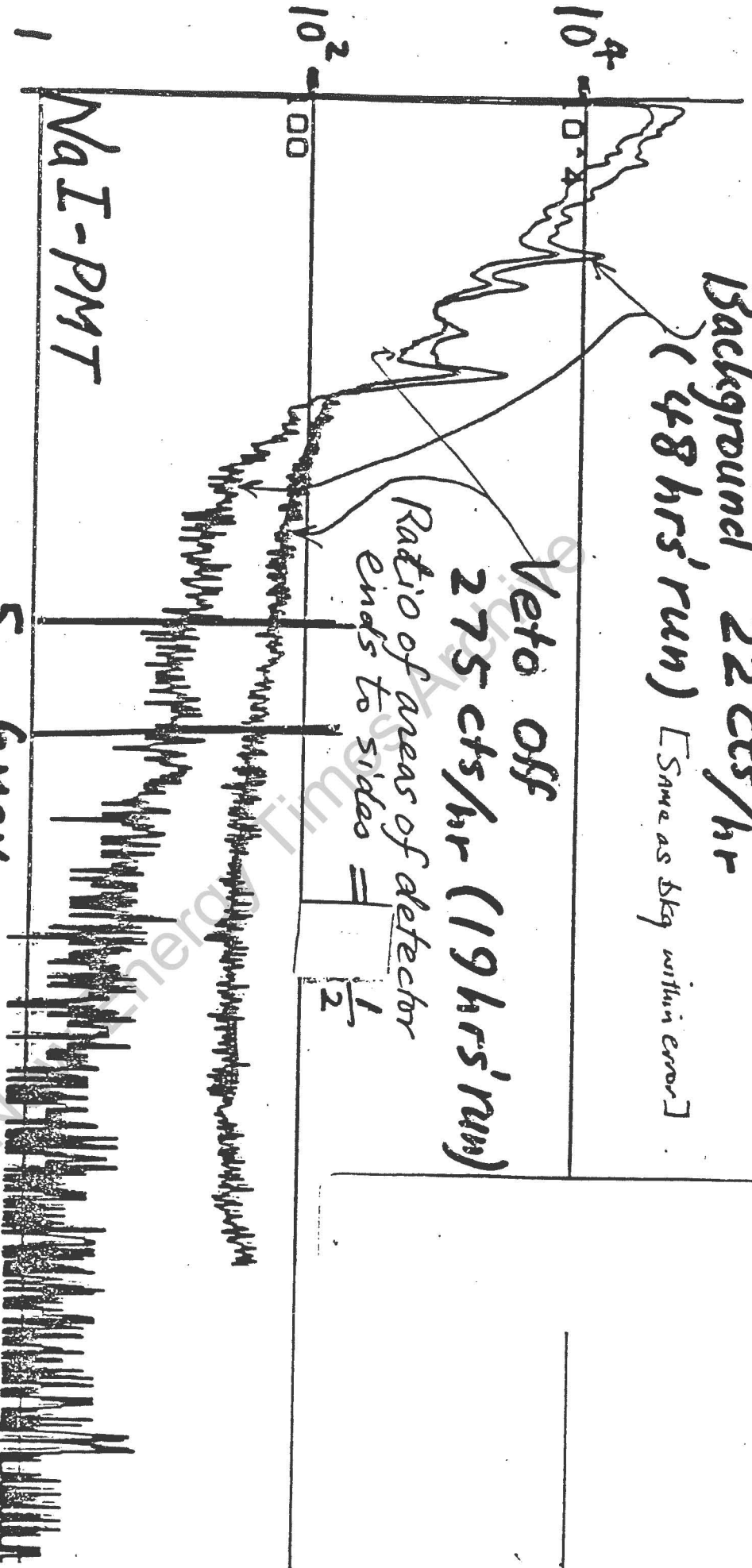
10<sup>6</sup> TO<sup>6</sup>B

Cast Pd sphere (4 mm dia)

Background 22 cts/hr  
(48 hrs' run) [Same as sky within error]

Veto off  
275 cts/hr (19 hrs' run)

Ratio of areas of detector  
ends to sides =  $\frac{1}{2}$



LC- 5.0023MeV  
SLC 78

6.0138MeV  
13

Figure 5

$R = Q_{out}/Q_{in}$   
(Heat Ratio)

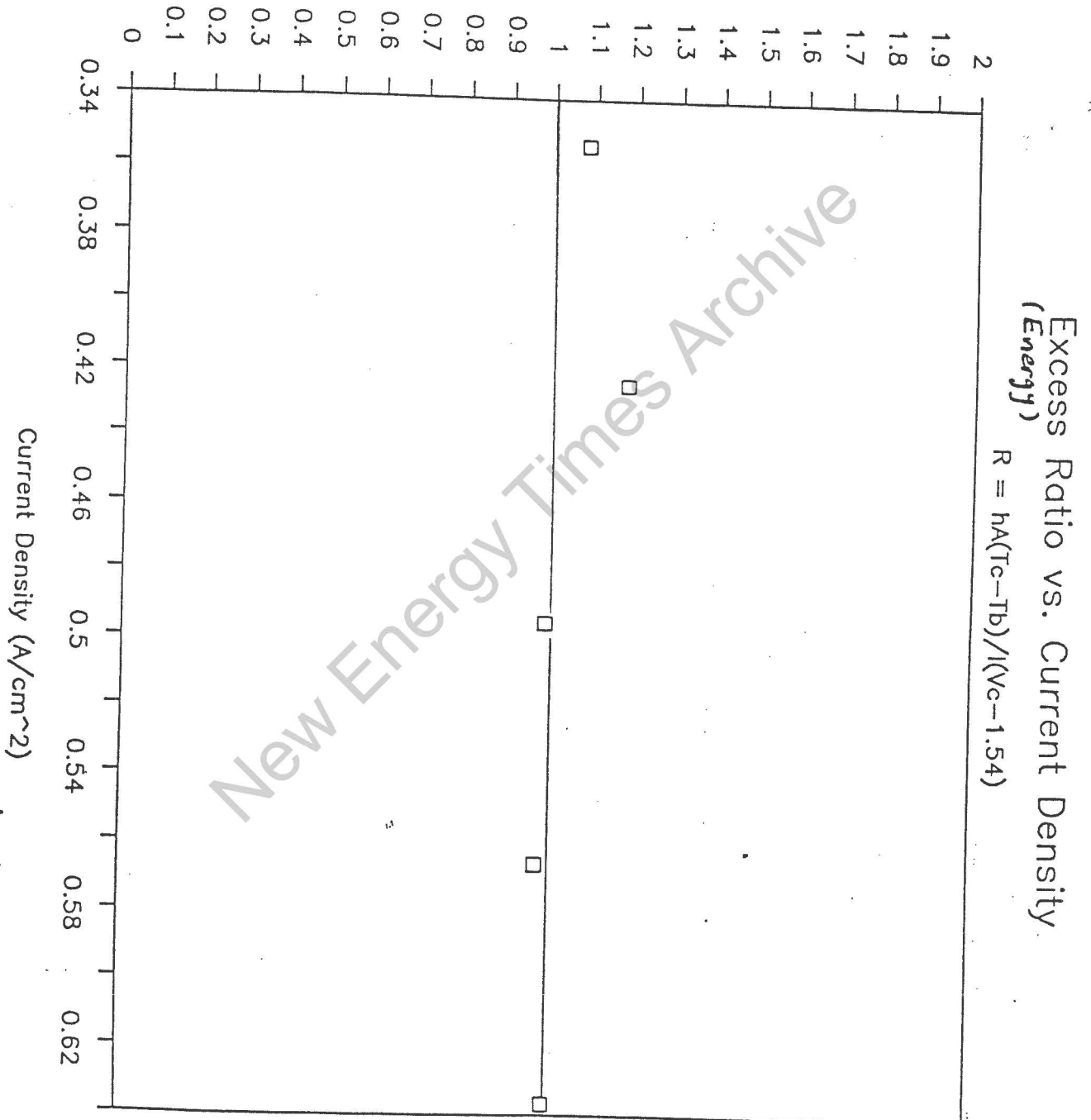


Figure 6